Biological Services Program

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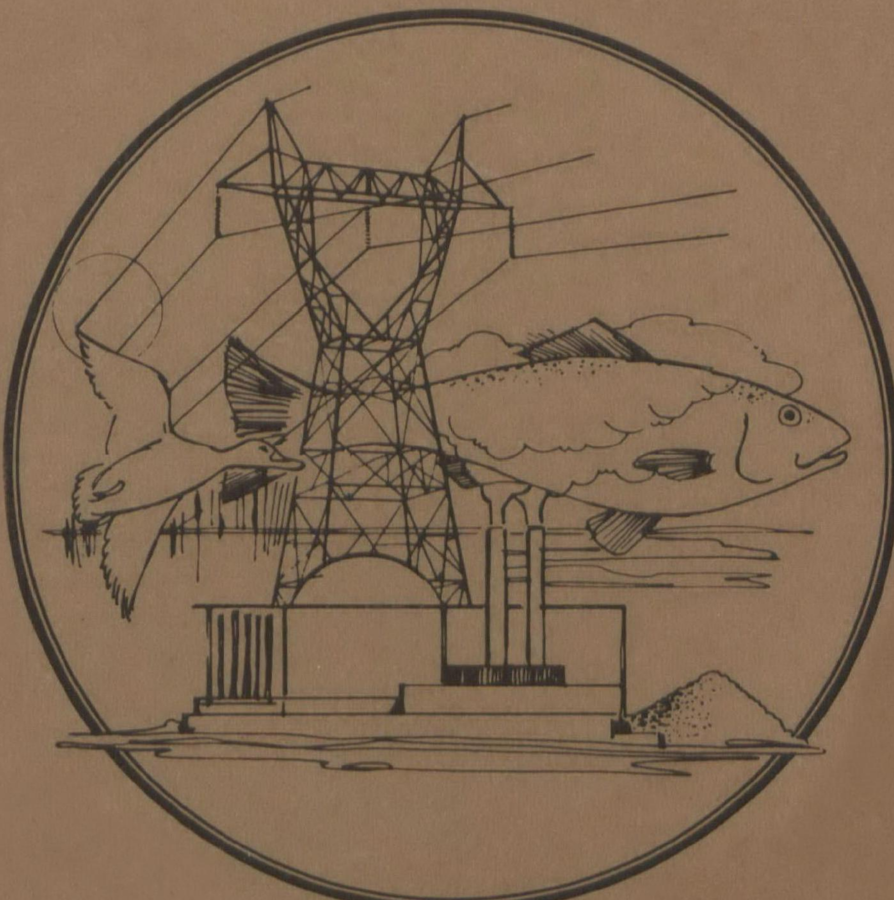
Air Pollution and Acid Rain,
Report No. 3

THE EFFECTS OF AIR POLLUTION AND ACID RAIN ON FISH, WILDLIFE, AND THEIR HABITATS

INTRODUCTION



Office of Research and Development
U.S. Environmental Protection Agency

Fish and Wildlife Service

U.S. Department of the Interior

The Biological Services Program was established within the U.S. Fish and Wildlife Service to supply scientific information and methodologies on key environmental issues that impact fish and wildlife resources and their supporting ecosystems.

Projects have been initiated in the following areas: coal extraction and conversion; power plants; mineral development; water resource analysis, including stream alterations and western water allocation; coastal ecosystems and Outer Continental Shelf development; environmental contaminants; National Wetland Inventory; habitat classification and evaluation; inventory and data management systems; and information management.

The Biological Services Program consists of the Office of Biological Services in Washington, D.C., which is responsible for overall planning and management; National Teams, which provide the Program's central scientific and technical expertise and arrange for development of information and technology by contracting with States, universities, consulting firms, and others; Regional Teams, which provide local expertise and are an important link between the National Teams and the problems at the operating level; and staff at certain Fish and Wildlife Service research facilities, who conduct inhouse research studies.

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UNITED STATES
DEPARTMENT OF THE INTERIOR
FISH AND WILDLIFE SERVICE

EASTERN ENERGY AND LAND USE TEAM
Route 3, Box 44
Kearneysville, West Virginia 25430

Dear Colleague:

The Eastern Energy and Land Use Team (EELUT) is pleased to present you the results and synthesis of available information on the effects of air pollution and acid rain or precipitation on fish, wildlife, and their habitats. This Introduction is the first in a series of nine reports relating acid rain and air pollution to forests, grasslands, lakes, rivers and streams, deserts and steppes, arctic tundra and alpine meadows, urban ecosystems, and critical habitats of threatened and endangered species. The series, "The Effects of Air Pollution and Acid Rain on Fish, Wildlife, and their Habitats" (FWS/OBS - 80/40.3, 40.4, 40.5, 40.6, 40.7, 40.8, 40.9, 40.10, 40.11) was prepared under the direction of David Adler at Dynamac Corporation, Rockville, Maryland.

This Introduction presents an overall view of the major air pollutants including photochemical oxidants, particulates, and acidifying air pollutants. The remaining series relate the effects of these pollutants to several major ecosystems in the United States.

Please feel free to send suggestions or comments to EELUT so that we may continually strive to improve our future products.

Sincerely,

Edgar A. Pash

Edgar A. Pash
Team Leader, EELUT

Enclosure

FWS/OBS-80/40.3
June 1982

AIR POLLUTION AND ACID RAIN, REPORT 3

THE EFFECTS OF AIR POLLUTION AND ACID RAIN ON FISH, WILDLIFE, AND THEIR HABITATS

INTRODUCTION

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DISCLAIMER

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ABSTRACT

Air pollution and acid rain impacts on living resources are a major source of concern to the U.S. Fish and Wildlife Service and other governmental agencies charged with the protection of natural resources and the environment. This introductory volume synthesizes the results of scientific research related to air pollution effects on fish and wildlife resources. It is intended for use as a general reference and to provide background information for the eight ecosystem specific reports in this series: Deserts, Forests, Grasslands, Lakes, Rivers and Streams, Tundra and Alpine Meadows, Urban Ecosystems, and Critical Habitats of Threatened and Endangered Species.

Air pollutants related to effects on fish, wildlife and their habitats are classified into three categories: photochemical oxidants, particulates, and acidifying air pollutants. A general summary of pollutant origins, atmospheric transport, transformation and deposition is presented in this volume. The bulk of this report describes plant, animal, and ecosystem responses to air pollution as well as factors affecting the sensitivity of receptive ecosystems. The Introduction also briefly describes relevant features of air quality legislation. A computerized bibliography and special reference library have been prepared in support of this series.

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1.0 INTRODUCTION

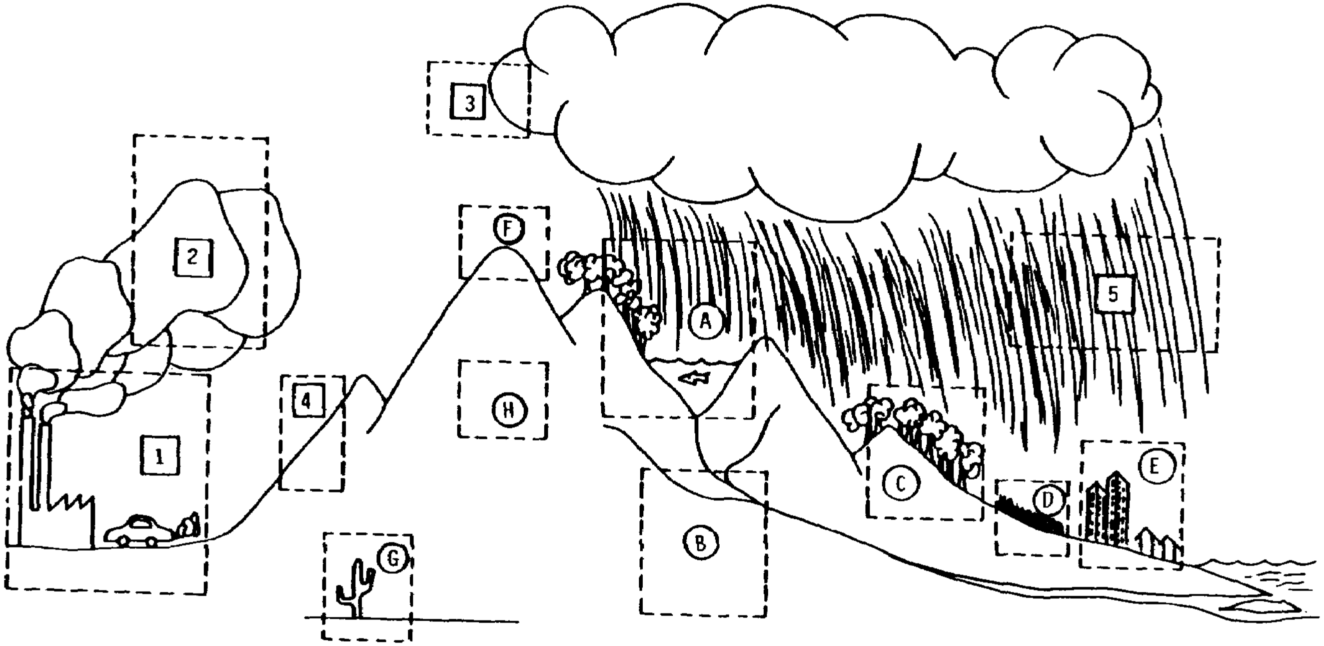
This introductory volume and accompanying reports taken together constitute a review of the state of current knowledge concerning the effects of air pollution and acid rain on fish, wildlife, and their habitats. The U.S. Fish and Wildlife Service (FWS), as a source of ecological expertise within the Department of Interior, acts as a focal point for research, methodological development, and dissemination of information to improve the effectiveness of decisionmaking in areas affecting natural resources. Particularly important are decisions which must weigh the advantages of development projects against potential long-term harm to the environment from resulting pollution.

The FWS has statutory responsibility to protect the nation's fish and wildlife resources and specifically to review and comment on the environmental impacts of proposed developments. This set of reports on air polution and related acid precipitation is intended to strengthen the ability of the FWS to meet this responsibility by providing a usable summary of what is known about the impacts and what is yet to be learned. The focus is on the effects of air pollution, the long-range transport of air pollution (LRTAP), and atmospheric deposition on fish and wildlife. The reports are general in nature with extensive reference to the technical literature provided for those requiring additional information beyond the scope of these documents.

The reports are the result of efforts to review and consolidate information from current research and the available literature into a format appropriate to FWS needs. They are primarily oriented toward the requirements of biologists and Federal and state land managers called upon to assess the ecological impacts of air pollution. However, as a synthesis of information it is expected that the reports will be of interest to a diverse audience including policymakers, public interest groups, consultants, and others concerned specifically with the anticipation and mitigation of detrimental effects of air pollution on wildlife resources.

This series consists of nine individual survey reports; the principal topics covered are depicted in Figure 1. This volume provides an introduction to the nature of air pollution, its sources, atmospheric transport, transformation, deposition and fate in terrestrial and aquatic ecosystems. The biological and ecological effects of air pollution, as well as factors affecting ecosystem sensitivity, are discussed. A brief introduction to air quality legislation is included. Also, a glossary of important terms can be found at the end of this report. The remaining reports in this series provide detailed information specific to effects within the eight ecosystems represented schematically in Figure 1. The series is supported by a computerized bibliography and a library of the scientific literature employed.

To keep the discussion within manageable bounds, a simplified classification of air pollution has been used which is consistent with the focus of the series on effects as observed in the field. Pollutants affecting wildlife resources are classified into one of three categories:



POLLUTANTS

Key: ☐ 1 - Air Emissions
2 - Atmospheric Pollutants
3 - Pollutant Transport and Transformation
4 - Dry Deposition
5 - Wet Deposition

ECOSYSTEMS

○ A - LAKES
B - RIVERS AND STREAMS
C - FORESTS
D - GRASSLANDS
E - URBAN ECOSYSTEMS

F - TUNDRA AND ALPINE MEADOWS
G - DESERTS
H - CRITICAL HABITATS OF THREATENED AND ENDANGERED SPECIES

Figure 1. Principal components of the air pollution/acid rain system in relation to the ecosystems addressed by this series.

- Photochemical oxidants. These pollutants constitute a large variety of substances, or "secondary" pollutants, which are produced by chemical reactions in the atmosphere as a result of sunlight acting on nitrogen oxides and hydrocarbons. These "primary" pollutants in turn may be the result of either natural or man-made processes. The most important impacts of photochemical oxidants are on terrestrial plants.

- Particulates and other toxic pollutants. Included in this category are the heavy metals, fluorides, and organic micropollutants. These can affect plants, animals, and the quality of their habitats in many different ways.

- Acidifying air pollutants. These pollutants share the characteristic that their effects on plants and animals are due to acidity. They may be acids such as those found in acid deposition or they may be gases, such as SO_2, which are converted to acids when incorporated into living systems.

Because the series addresses the needs of the FWS, the emphasis of the reports is on the effects of air pollution as observed in the field. In general, there are three types of effects:

- abiotic effects on air, land, and water such as leaching phenomena resulting in the movement of nutrients or changes in the acidity of surface water;

- aquatic and terrestrial biotic effects such as increased susceptibility of plants to disease and predation or impacts on flora and fauna resulting from changes in water or soil chemistry; and

- ecosystem effects including complex interactions between biotic and abiotic components exemplified by forest ecosystems in which decreased productivity of trees and increased nutrient mobility caused by air pollution can cause changes in evapotranspiration, microclimate and other factors, ultimately leading to large losses of nutrients from the ecosystem and subsequent effects on downstream aquatic ecosystems.

These effects may be acute, episodic and short-term, or they may be chronic, cumulative and long-term. A great deal of further study is needed to assess their potential reversibility.

Each of the ecosystem-specific reports in this series contains a brief background discussion of biotic, abiotic, and functional ecosystem aspects relevant to an assessment of air pollution impacts. The current state-of-knowledge concerning biological and ecological impacts of air pollution is introduced for each ecosystem and related socio-economic considerations are briefly outlined where appropriate.

The final chapter of each ecosystem-specific report discusses topics for further research related to the specific ecosystems. Some research areas closely complement FWS experience and capability while others are beyond the scope of the agency. As a general rule, these sections point out that an effective evaluation of air pollution effects will require further investigations in established disciplines of plant and animal physiology, plant pathology and the biomedical sciences. The complexity of effects stemming from phenomena such as LRTAP, the long-range transport of air pollutants (especially acids, oxidants, trace elements, and the organic micropollutants), poses significant challenges to research efforts in these fields. In view of the expense and specialization required to develop knowledge of these subjects, FWS activities in the following areas appear especially useful:

- a continuing synthesis and dissemination of the literature on plant and animal effects in specific ecosystems as it relates to the needs of FWS personnel;

- the creation of a working relationship between key researchers or research institutions and the FWS personnel who rely on the development of specific areas of knowledge or expertise; and

- efforts to expand the use of wildlife species and selected plants on which they depend in field experimentation related to the biotic effects of air pollutants and LRTAP.

Several reports point to the potential utility of indicator plants and animals in pollution-related research applications. In view of the FWS role in wildlife protection, habitat preservation and the development of baseline ecological knowledge to support these activities, biomonitoring research would appear to be a feasible and potentially productive activity complementing other agency responsibilities. Furthermore, as the monitoring of ambient air pollution and precipitation chemistry is expanded to remote areas of the country, biomonitoring undertaken by field specialists may be one of the more practical methods of discerning potential biotic effects in these remote areas.

Increased understanding of the complex impacts at the ecosystem level will not be easily or quickly achieved. Again, a continuing synthesis and dissemination of relevant ecosystem-specific literature is important. The ecological research should be complemented by research related to the social, economic, and intergenerational aspects of large-scale alterations in ecosystem functioning. Although such studies have a high degree of uncertainty associated with them, they are necessary for a complete picture of the impacts of air pollution and acid deposition on fish, wildlife, and their habitats.

2.0 PRINCIPAL CATEGORIES AND ORIGINS OF MAJOR AIR POLLUTANTS

For the purposes of this series of reports the air pollutants of greatest significance for potential impact to fish, wildlife and their habitats will be classed into three functional categories:

- photochemical oxidants;
- particulates; and
- acidifying air pollutants.

This classification is useful to a review of biological effects since the individual pollutants in each category possess similarities in physical and chemical properties and cause similar effects. The pollutants of a class also share similar atmospheric dispersion and deposition processes, follow common exposure pathways to wildlife and habitat, and elicit like responses among the different components of the ecosystem. Some pollutants of less significance to fish and wildlife effects may fit only loosely within this functional categorization.

Air pollutants may be emitted from point sources (fixed identifiable sources) or area sources (numerous point or mobile sources such as chimneys or automobiles). Pollutants from point and area sources are referred to as primary pollutants. Interactions among two or more primary pollutants and normal atmospheric constituents create secondary pollutants. Many of these chemical reactions require photoactivation. The rates, reaction routes, and intermediate steps involved in the process are influenced by many factors such as relative concentration of reactants, degree of photoactivation, variable meteorological dispersive forces, influences of local topography, ambient temperature and relative humidity.

Although air pollution is usually associated with human activities, many of the major gaseous pollutants also have natural sources. Taken on a worldwide basis, natural emissions of some pollutants, such as nitrogen oxides from volcanic eruptions, often exceed anthropogenic, or man-made, emissions by several orders of magnitude (Urone 1976). The reason man-made emissions so noticeably affect the quality of the environment is that they tend to concentrate locally or regionally in air masses downwind of urban areas or large point sources.

2.1 PHOTOCHEMICAL OXIDANTS

The group of pollutants known as photochemical oxidants are secondary pollutants, that is, they are produced by complex chemical reactions between primary pollutants, notably nitrogen oxides and hydrocarbons. These reactions are common in the atmosphere above urban centers where there are hundreds of different hydrocarbons. Because of the multitude and variety of photochemical oxidants, very few individual oxidants have been observed. Table 1 lists the major oxidants which are of primary concern as a pollution problem (National Research Council 1977). These are generally found in association with other photochemical products, including aldehydes, formic acid, nitrous acid, gaseous and particulate nitrates, and sulfates.

Table 1. Major pollutants of the photochemical oxidant complex.

Pollutant	Formula
Ozone	O_3
Peroxyacetylnitrate (PAN)	$CH_3COO_2NO_2$

Ozone (O_3) is by far the most abundant of the photochemical oxidants. It is a product of many different reactions, and also occurs naturally in large quantities in the stratosphere (Singh et al. 1980). Peak concentrations in urban and suburban areas may range from 0.3 to 0.6 ppm while peak ozone levels in rural areas seldom exceed 0.2 ppm (U.S. EPA 1978a). Average concentrations in rural areas may often equal or surpass urban averages due to pollutant transport and the regional scale of oxidant pollution. Averaged ambient ozone levels in urban areas showed no long-term trend over the period from 1974 to 1979, however national averages were observed to decline by 3 percent between 1978 and 1979 (USEPA 1980a).

Peroxyacetylnitrate (PAN) is actually the most abundant member of a series of similar compounds, but it appears to be the only one found to occur in photochemical smog at concentrations high enough to pose a pollution problem. Rural daily maximums rarely exceed 3.0 ppb, however maximum urban valves can reach levels of 0.05 to 0.2 ppm (USEPA 1978a). PAN is known to be of greater phytotoxicity than ozone; however elevated levels of ozone are more common, thus ozone is considered to be the most injurious of the photochemical oxidants.

The precursors of photochemical oxidants are numerous, but generally fit into the categories of nitrogen oxides (NO_x) or hydrocarbons (HC). These pollutants are emitted in large quantities as a result of fossil fuel combustion. Estimated national emissions of these precursors are presented in Table 2 by source.

The nitrogen oxide precursors of the photochemical oxidant complex figure prominently among the acidifying air pollutants and will be described in detail in Section 2.3. Major types of hydrocarbon precursors found in urban atmospheres include the alkenes, alkynes, cycloalkanes and aromatics. Hydrocarbons, as a group, constituted the second largest form of air pollution by mass in the U.S. in 1977 (USEPA 1978b).

Table 2. Estimated national anthropogenic emissions of photochemical oxidant precursors in 1977.

Pollutant Source	Annual Production (10^6 metric tons/year)	
	NOx	HC
Transportation	9.2	11.5
Fuel Combustion (Stationary Sources)	13.0	1.5
Industrial Processes	0.7	10.1
Solid Waste Disposal	0.1	0.7
Miscellaneous	0.1	4.5
Totals	23.1	28.3

(From USEPA 1978b)

Transportation and stationary source fuel combustion (e.g., power plants) are the principal sources of the oxides of nitrogen (USEPA 1978b). Solid waste disposal, industrial process losses, and miscellaneous releases account for the remaining NO_x emissions. Almost all anthropogenic NO_x is produced by the oxidation of atmospheric nitrogen (N_2) at high temperature during combustion. Hence, the formation of NO_x is often promoted under those operating conditions where the combustion of fuel is optimized (Stoker and Seager 1976). Figure 2 presents estimated trends in gross and net NO_x emissions, by source, through the year 2000.

Natural sources of NO_x contribute significantly more than human-related sources (Stoker and Seager 1976, Soderlund and Svensson 1976). Bacterial action in soils, resulting in the decomposition of nitrogen-containing compounds, represents the major natural source of nitrous oxide (N_2O), some 592 million tons (Stoker and Seager 1976). Bacterial activity is also responsible for an estimated annual production of 430 million tons of nitric oxide (NO). Lightning also produces NO_x but the amount is negligible.

The highest average concentrations of nitrogen oxides are found in heavily populated, industrialized urban areas. Average ambient levels greater than 1.0 ppm NO_x are seldom encountered in cities, while rural air usually averages a few parts per billion (USEPA 1978a). One-hour maximum concentrations of 2.5 and 4.5 ppm nitrogen dioxide (NO_2) have been

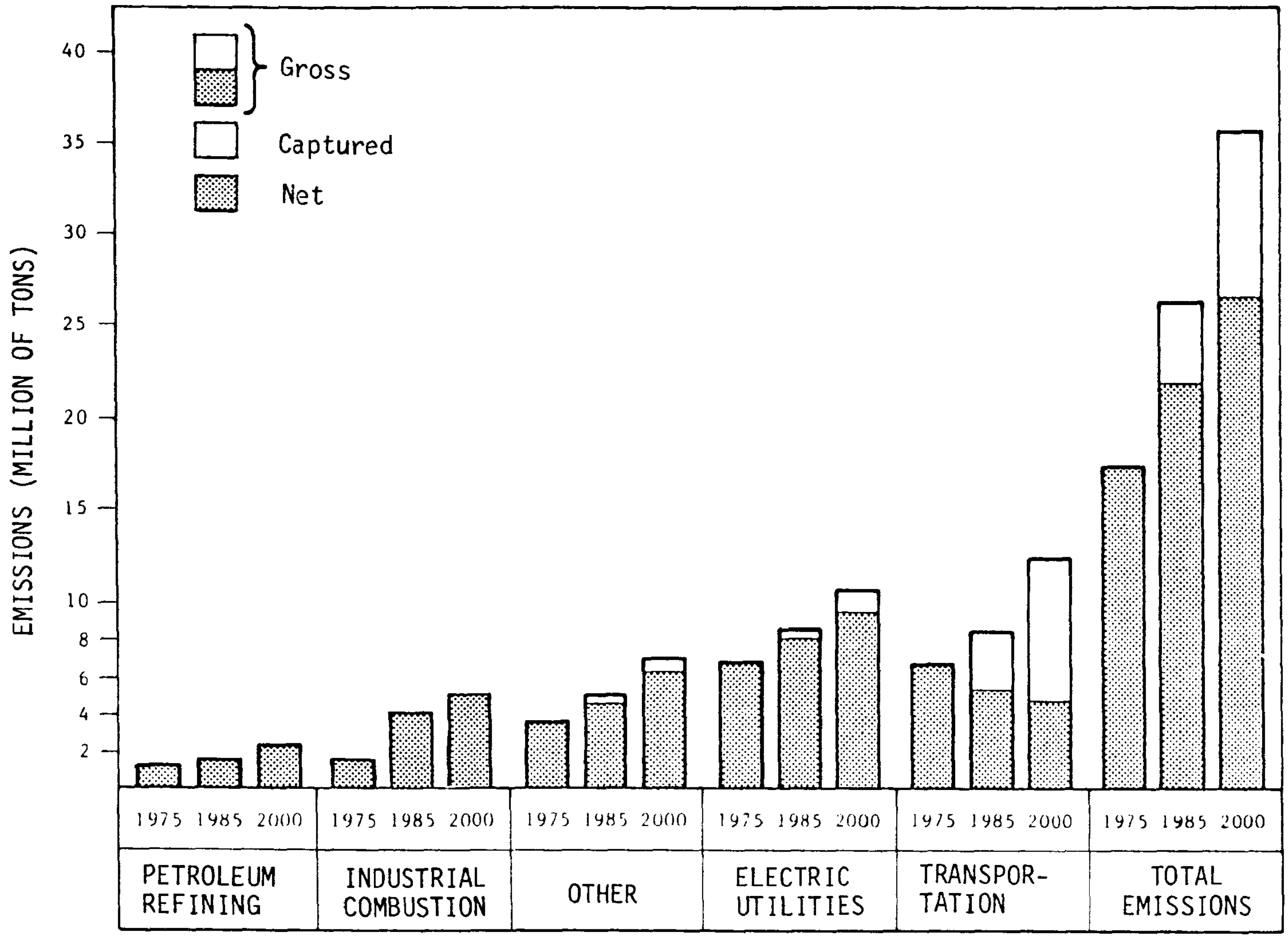


Figure 2. Estimated trends in anthropogenic nitrogen oxide emissions, by source (high growth scenario). (From USEPA 1980b)

recorded in Chicago and Philadelphia, respectively, whithout exceeding the primary air quality standard, set by the U.S. Environmental Protection Agency, of 0.05 ppm NO_2 on an annual average; levels in excess of this standard are consistently found in the Los Angeles area (National Research Council 1977). Due to projected increases in transportation and stationary source emissions, observations of nitrogen dioxide concentrations showed an increasing trend between 1974 and 1979 (USEPA 1980a).

Transportation and industrial processes, particularly refining, are the principal emitting sources of hydrocarbons. Automobile combustion alone accounts for half of the total anthropogenic emissions of hydrocarbons. Other large sources include solvent evaporation and hydrocarbon emissions resulting from various chemical manufacturing processes (Clark 1980; Tilton and Bruce 1980). Trends in gross and net hydrocarbon emissions, by source, are given through the year 2000 in Figure 3.

Natural sources of hydrocarbons are also important. In 1968, natural sources of hydrocarbons contributed 85 percent of the total atmospheric input (Stoker and Seager 1976). The two principal types of natural emissions are methane (CH_4), and a group of organics known as terpenes. Copious amounts of methane are generated in swamps, marshes, and similar water bodies as a result of the anaerobic bacterial decomposition of organic matter. Methane does not contribute to photochemical oxidant formation (Tilton and Bruce 1980). The terpenes, a family of high molecular weight hydrocarbons, are produced by vegetation. About 43 percent of these emissions occur in the summer months, and about 45 percent of the annual emissions occur in the southern United States (Tilton and Bruce 1980).

On a nationwide scale, there is a direct correlation between population density and ambient hydrocarbon levels. The areas with the highest density of non-methane hydrocarbon emissions are: 1) the northeast corridor; 2) certain industrial states (Ohio, New York, Pennsylvania, Illinois, and Indiana); and 3) states along the gulf coast where petroleum refining and storage activities contribute heavily to hydrocarbon emissions (Tilton and Bruce 1980).

Non-methane hydrocarbon concentrations typically average 1.0 ppm in urban air and less than 0.1 ppm in rural air, however levels as high as 10.0 ppm have been recorded in Los Angeles (USEPA 1978a). National hydrocarbon emissions decreased by 4 percent in the period from 1970 to 1979, primarily due to the achievement of lower emissions from transportation sources (USEPA 1980a).

2.2 PARTICULATES

Particulate matter in the atmosphere exists in a wide variety of forms, usually as smoke, fumes, mists, oils, and dusts (Anon. 1969). Virtually all of these pollutants fall into the category of aerosols: solid particles or liquid droplets which are dispersed or suspended in a gaseous

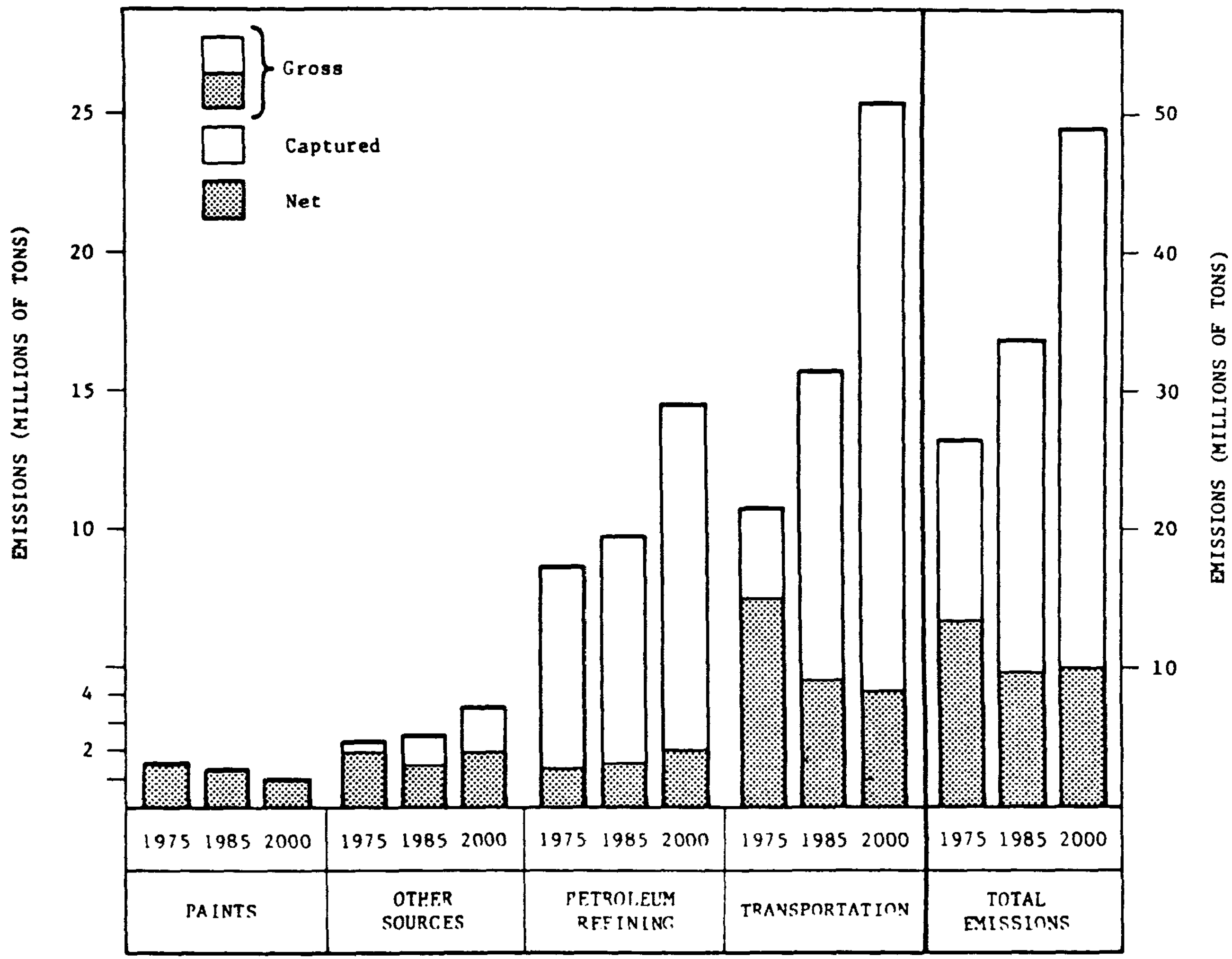


Figure 3. Estimated trends in anthropogenic hydrocarbon emissions, by source (high growth scenario). (From USEPA 1980b).

medium. They may range in chemical composition from a single elemental species to highly complex substances incorporating virtually any of the other atmospheric pollutants. Common components of atmospheric particulate burdens are listed in Table 3.

The specific composition of particulates greatly depends on the type of emission source, industrial operating conditions, characteristics of fuels employed, and the nature of emission-control equipment. In view of its chemical diversity, particulate matter is usually categorized according to its size. Atmospheric particles generally range in size from molecular clusters (< 0.005 um) to visible dust (> 100 um). Within this range, they are classified as coarse or fine particles, depending on their diameter and mode of formation. The primary particulates are generally coarse (> 2.0 um) and are emitted directly from anthropogenic and natural sources. The majority of fine particles (< 1.0 um) in the atmosphere are secondary particulates that result from chemical reactions and transformations among ambient air pollutant mixtures (Fennelly 1976).

In industrialized regions, sulfates ($SO_4^{=}$) are by far the most abundant of the fine particulates (Fennelly 1976). Under natural conditions, more sulfate aerosols are associated with ammonium ions (NH_4^{+}) than exist as sulfuric acid (H_2SO_4), however sulfuric acid and compounds such as lead sulfate ($PbSO_4$) predominate in urban air. Nitrate ions (NO_3^{-}) are common in both gaseous and particulate form in rural and urban air (Galloway et al. 1981). These acidifying pollutants are discussed in the following section.

Carbonaceous aerosols are also abundant in fine particulate matter. These usually contain an elemental component, often graphite or soot, and an organic component, usually a hydrocarbon (Miller et al. 1979). Trace organics include the polychlorinated biphenyls (PCBs), chlorinated hydrocarbon pesticides (HCCls) and polycyclic aromatic hydrocarbons (PAHs) (Galloway et al. 1981). Such substances may exist as solid entities or they may adsorb to other particulates from a vapor phase.

Atmospheric metals and trace elements range widely in size, and are usually present in the solid form. Those found in a vapor phase include arsenic (As), cadmium (Cd), mercury (Hg), and selenium (Se) (Miller et al. 1979; Galloway et al. 1981). Most of the metals combine with oxygen, sulfur, sulfate, or nitrate following their release to the atmosphere, and are virtually always found in association with various kinds of particulate matter (Linton et al. 1976).

Particulates of lesser abundance include a variety of radioactive particles, the chief among them being isotopes of uranium (^{235}U, ^{238}U); radioactive gases such as radon (^{222}Rn) may adsorb to particulates (Rivera-Cordero 1970). Nutrient elements such as nitrogen (N), phosphorus (P), and potassium (K) also exist in a variety of forms as atmospheric particulates. Finally, there are many inorganic particles of natural origin, including siliceous dusts, sea salts, and carbonates which contribute to atmospheric loadings of particulate matter.

Table 3. Chemical constituents of particulate pollution.

Non-metallic Ions

- Sulfate ($SO_4^{=}$)
- Ammonium (NH_4^{+})
- Nitrate (NO_3^{-})[a]
- Chloride (Cl^{-})
- Fluoride (F^{-})

Nutrient Elements

- Nitrogen (N)
- Potassium (K)
- Phosphorus (P)

Radioactive Particles

- Uranium (^{235}U, ^{238}U)

Synthetic Organic Compounds[a]

- Polycyclic Aromatic Hydrocarbons (PAH)
- Chlorinated Hydrocarbons (HCCl)
- Polychlorinated Biphenyls (PCB)

Natural Organic Dust

- Pollen, spores, fungal hyphae and plant parts

Trace Elements

- Aluminum (Al)
- Antimony (Sb)
- Arsenic (As)[a]
- Beryllium (Be)
- Cadmium (Cd)[a]
- Chromium (Cr)
- Cobalt (Co)
- Copper (Cu)
- Iron (Fe)
- Lead (Pb)
- Nickel (Ni)
- Manganese (Mn)
- Mercury (Hg)[a]
- Molybdenum (Mo)
- Selenium (Se)[a]
- Silver (Ag)
- Thallium (Tl)
- Tin (Sn)
- Titanium (Ti)
- Vanadium (V)
- Zinc (Zn)

[a]These substances may also be present in a vapor phase

The primary anthropogenic sources of atmospheric particles are stationary fuel combustion and industrial processes (USEPA 1978b). Industrial sources include non-ferrous metal smelters, iron and steel mills, petroleum refineries, cement quarries, pulp and paper plants, asphalt and chemical works, and manufacturers of soap and synthetic detergents, glass and glass fiber, and textiles (Anon. 1969). Other human activities generating particulate pollution include the cultivation and fertilization of agricultural and forest land, solid waste disposal, commercial and residential heating, and the operation of motor vehicles. Table 4 quantifies national particulate emissions according to various types of sources.

Table 4. Estimated national anthropogenic emissions of particulates in 1977.

Pollutant Source	Annual Production (10^6 metric tons/year) Particulates
Transportation	1.1
Fuel Combustion (Stationary Sources)	4.8
Industrial Processes	5.4
Solid Waste Disposal	0.4
Miscellaneous	0.7
Total	12.4

(From USEPA 1978b)

The majority of fine ambient aerosols result from the incomplete combustion of fossil fuels and from residuals produced during incineration, manufacturing, photochemical, and condensation processes (Lee 1972). Greater amounts of fine particles are known to be emitted from oil-fired burners than from coal-fired units (Cheng et al. 1976). Moreover, the finer particulates from fossil fuel combustion have been found to preferentially concentrate toxic trace elements on the particle surface (Linton et al. 1976). Toxic vapor-phase contaminants, such as the polycyclic aromatic hydrocarbons and volatile metals, adsorb preferentially to fine particulates of respirable size (Miller et al. 1979).

The natural sources of gaseous pollutants often generate particulate matter and trace metals as well. Volcanic activity is one of the important geochemical origins of these substances; another is wind erosion (Peirson et al. 1973). Sea spray is the major marine source and is an important localized contributor of atmospheric aerosols in coastal areas. The principal sources of particulate matter related to biota include forest and grassland fires, wind-blown bacteria, and biological releases such as plant pollen and the spores of fungi (National Atmospheric Deposition Program 1978). Atmospheric metals also originate in vapor emissions from land, the seas, and vegetation, including the process of low-temperature volatilization from soils (Galloway et al. 1981). Historic base levels of naturally-produced particulates are very often dwarfed by anthropogenic emissions which dominate the air pollution situation in certain regional and localized contexts. Table 5 illustrates this point by comparing ratios of anthropogenic to natural emissions of the predominant metal particulates on a global scale. The ratios, or mobilization factors, may be significantly greater than these averages in large portions of the United States since emission sources are much more concentrated than in remote portions of the globe (Galloway et al. 1981).

Regional concentrations of atmospheric particulates vary widely throughout the United States. Variations in ambient levels for different trace metals are shown in Table 6, which presents median levels of atmospheric trace metals in remote, rural, and urban areas. Particulate concentrations also fluctuate both diurnally and seasonally. Levels of vanadium and nickel, for example, correlate with the use of fuel oils for heating and are thus present in air in greatest quantities during winter (Galloway et al. 1981). Submicron aerosols are found to predominate in urban and suburban areas while larger particles comprise the majority of rural and remote samples (Miller et al. 1979; Van Vaeck et al. 1979).

Particulate emissions declined nationally by 50 percent between 1970 and 1979, largely due to controls on industrial and utility emissions, decreased burning of solid waste, and reduced coal burning by small sources (USEPA 1980a). As a result, ambient levels have shown a consistent downward trend over this period. Evidence suggests, however, that local concentrations of particulates in the fine or respirable range are generally increasing due to the dispersal of point sources from urban areas. For example, visibility degradation in the southwest and eastern United States is observed to be spreading as a result of gradual shifts in particle size distributions toward the finer, light-scattering sizes (USEPA 1980a, b). Estimated trends in net particulate emissions, by source through the year 2000, are given in Figure 4. These projections also suggest that, despite controls, total atmospheric particulate burdens may increase due to the growing number of sources.

Table 5. Estimated global emissions of atmospheric metals from natural and anthropogenic sources. Ratios of anthropogenic to natural emissions provide mobilization factors.

Metals	Emissions (10^8 g/yr)		Mobilization Factors (Ratio of Anthropogenic to Natural Emissions)
	Natural	Anthropogenic	
Ag	0.6	50	83
As	29 [210][a]	780	3.3
Cd	2.9	55	19
Co	70	50	0.71
Cr	580	940	1.6
Cu	190	2,600	13
Hg	0.4 [250][a]	110	0.44
Mn	6,100	3,200	0.53
Mo	11	510	45
Ni	280	980	3.5
Pb	59	20,000	340
Sb	9.8	380	39
Se	4.1 [30][a]	140	4.7
Sn	52	430	8.3
V	650	2,100	3.2
Zn	360	8,400	23

[a]Volatile-phase emissions

(From Galloway et al. 1981)

Table 6. Average ambient trace metal concentrations in remote, rural, and urban atmospheres. Ratios indicate factors of concentration of urban and rural to remote ambient levels.

Metal	Average Ambient Concentrations (ug/m^3)			Ratios	
	Remote	Rural	Urban	Urban/Remote	Rural/Remote
Ag	0.01	0.3	1.1	110	30
As	0.2	6	25	125	30
Be	--	0.023	0.14	-	-
Cd	0.1	1.0	2.0	20	10
Co	0.05	0.1	10.0	200	2
Cr	0.3	5.0	40.0	133	17
Cu	0.2	6.0	100	500	30
Hg	0.5	2.0	20	40	4
Mn	0.4	30.0	150	375	75
Mo	0.3	-	2	7	-
Ni	0.36	2	30	83	6
Pb	1.0	100	2000	2000	100
Sb	0.2	3	30	150	15
Se	0.1	1.5	4.7	47	15
V	1.0	5	50	50	5
Zn	0.5	100	1000	2000	200

(From Galloway et al. 1981)

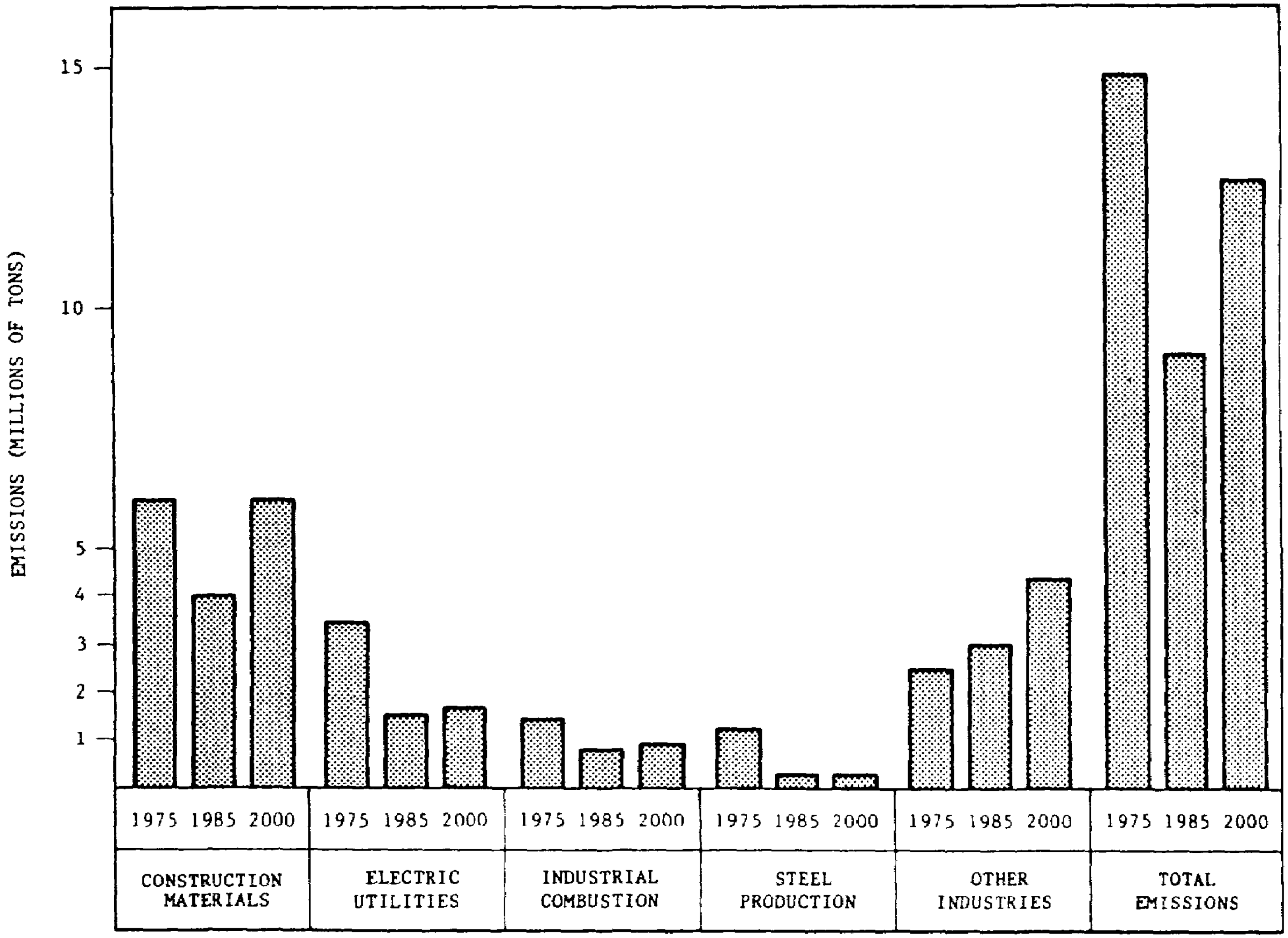


Figure 4. Estimated trends in net anthropogenic particulate emissions, by source (high growth scenario). (From USEPA 1980b)

2.3 ACIDIFYING AIR POLLUTANTS

The acidifying air pollutants include primary gaseous emissions associated with fossil fuel combustion and derivative acids resulting from the long-range transport and atmospheric transformation of these precursors. Table 7 presents the air pollutants most often implicated in the occurrence of local (primary pollutant) and regional (secondary pollutant) acidity effects in ecosystems.

Table 7. Primary and secondary forms of acidifying air pollutants.

Primary	Secondary
Sulfur oxides (SO_x)	Sulfuric acid (H_2SO_4)
Nitrogen oxides (NO_x)	Nitric acid (HNO_3)
Hydrochloric acid (HCl)	

Sulfur forms a number of oxides (SO_2, S_2O_3, SO_3, S_2O_7) but only sulfur dioxide (SO_2) and sulfur trioxide (SO_3) are important as gaseous air pollutants (Urone 1976). Usually only a small amount of SO_3 accompanies SO_2 (1 or 2% of the SO_2) and collectively the two are designated SO_x (Stoker and Seager 1976). The nitrogen oxides (NO_x) are composed of nitric oxide (NO) and nitrogen dioxide (NO_2).

These primary pollutants are transformed by atmospheric reactions to their corresponding acids and subsequently deposited, often hundreds of kilometers from their source, through wet or dry removal processes. Wet deposition is generally referred to as acid precipitation and accompanies episodes of rain, snow, hail, sleet, and fog. The dry deposition of atmospheric acids is also substantial, particularly during dry spells, although difficulties in the measurement of this component preclude a full understanding of its relative contribution to total acid deposition. Pollutant transport, transformation, and deposition processes are discussed further in Chapter 3.0.

Acid precipitation is defined as rain and related events of pH less than 5.6, a reference value for atmospheric water vapor in equilibrium with naturally occurring concentrations of carbon dioxide (CO_2) and its derivative carbonic acid (H_2CO_3) (Cogbill and Likens 1974; Galloway 1979; Hales 1980). The relation of acid rain to the logarithmic pH scale is depicted in Figure 5. Acid precipitation events are characterized by elevated concentrations of dissociated hydrogen ions (H^+) which results in lowered measured pH values.

The increased acidity of precipitation was first observed on a regional scale in Scandinavia, and attributed to the long-range transport of air pollutants from the industrial centers of Europe and the United Kingdom (Barrett and Brodin 1955; Bolin et al. 1971; Brosset 1973). Acid precipitation was soon documented in the northeastern United States (Likens et al. 1972; Cogbill and Likens 1974; Likens and Bormann 1974a; Galloway et al. 1976; Likens 1976), and subsequently in Florida (Brezonik et al. 1980) and in many regions of the west (Powers and Rambo 1981), including the states of Washington (Harrison et al. 1977; Dethier 1979), California (McColl and Bush 1978; Liljestrand and Morgan 1978), and Colorado (Lewis and Grant 1980a).

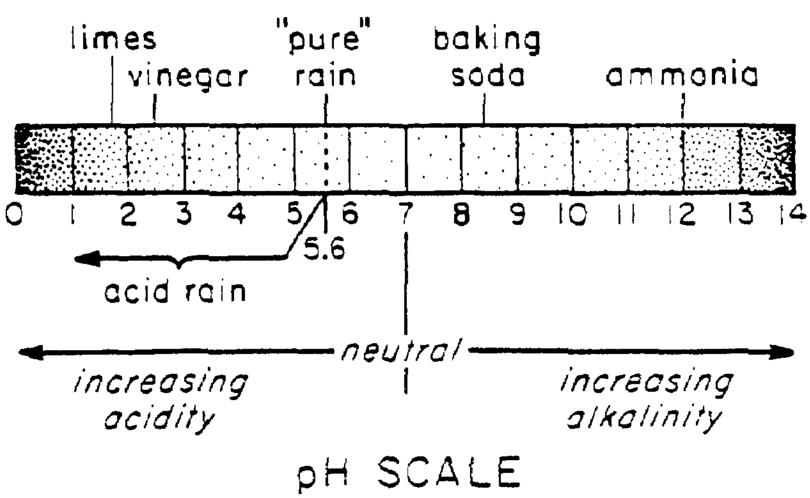


Figure 5. The pH scale with comparisons of acid rain to common acid and alkaline substances. Below pH 7.0, a unit decrease in pH is equivalent to a ten-fold increase in hydrogen ion concentration. (From Glass 1979a).

The primary constituents of acid precipitation are sulfuric acid ($H SO_4$) and nitric acid (HNO_3), although hydrochloric acid (HCl) and a variety of organic acids are minor contributors to precipitation acidity (Likens 1976; Likens et al. 1979; Galloway and Likens 1981). The average composition of strong acids in precipitation of the northeastern United States is shown in Figure 6.

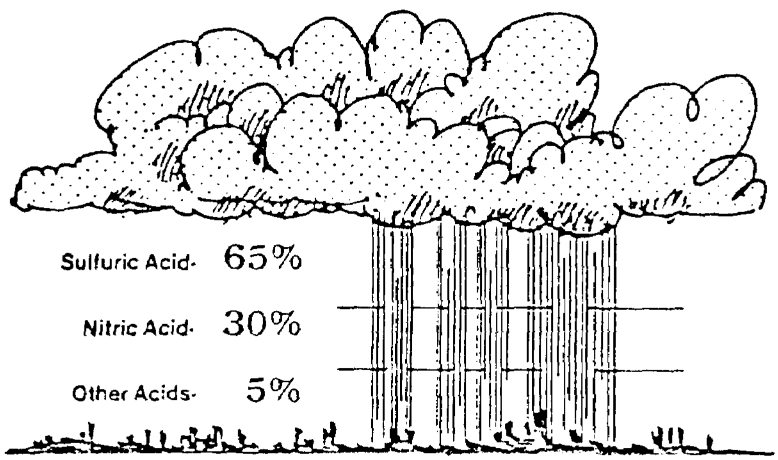


Figure 6. Typical northeastern U.S. acid rain components averaged annually. (From USEPA 1980c).

Significant geographic variations are found to occur in the relative composition of strong acids in precipitation. Sulfuric acid is observed to be the dominant anion in precipitation of the northeastern and northwestern United States (Likens 1976; Dethier 1979). In California and Colorado, the sulfuric acid fraction is less significant and the nitric acid component can assume values of 50 to 80 percent of total acids in precipitation (Lewis and Grant 1980a; McColl 1980; Morgan and Liljestrand 1980). The nitric acid component is increasing steadily in the northeastern United States, particularly in urban areas (Barnes 1979; Likens et al. 1979; Brezonik et al. 1980). From 1964 to 1979, in a calibrated watershed of New Hampshire, the importance of H_2SO_4 declined 30 percent relative to HNO_3 while the contribution of HNO_3 to acidity increased 50 percent relative to H_2SO_4 (Galloway and Likens 1981).

There are also seasonal variations in precipitation chemistry. For example, in New Hampshire the maximum contribution of H_2SO_4 to acidity observed was 73 percent in summer and 59 percent in winter, whereas HNO_3 contributed at most 31 percent in summer and 61 percent in winter (Galloway and Likens 1981). In the northeastern United States, the nitric acid fraction is more abundant in precipitation during winter than summer (Galloway 1979). The pH of precipitation is also consistently lower in summer than in other seasons of the year (Lioy 1979; Wolff et al. 1979).

The average annual pH of precipitation in industrialized regions of Europe and North America lies within the range of pH 4.0 to 4.5, and is reported to average pH 4.1 in the northeastern United States (Likens 1976; Likens et al. 1979; Hendrey and Lipfert 1980). Individual storms may be considerably more acidic than this average and some storms have been reported in the range of pH 2.1 to 3.5 (National Atmospheric Deposition Program 1978). Figure 7 gives the weighted mean pH of precipitation over the continental United States, for the years 1976 to 1979, from a compilation of data obtained through eleven precipitation chemistry monitoring networks. In some cases precipitation may be alkaline; pH averages above 6.0 have been reported in prairie regions where large amounts of alkaline dust are generated by winds (Cooper et al. 1976).

Recent efforts to improve ambient air quality by constructing tall stacks to discharge gaseous and particulate emissions have resulted in an increase in the residence time of sulfur and nitrogen oxides in the atmosphere. With this increased residence time comes a greater potential for acidifying transformations and a much larger area over which the pollutants can be deposited. For this reason it is difficult, if not impossible, to associate elevated levels of regional precipitation acidity with specific pollutant emission sources.

In the United States, sulfur oxides are released primarily from fossil fuel combustion (USEPA 1978b). Nitrogen oxide emissions, discussed earlier as precursors to photochemical oxidants, are produced during the

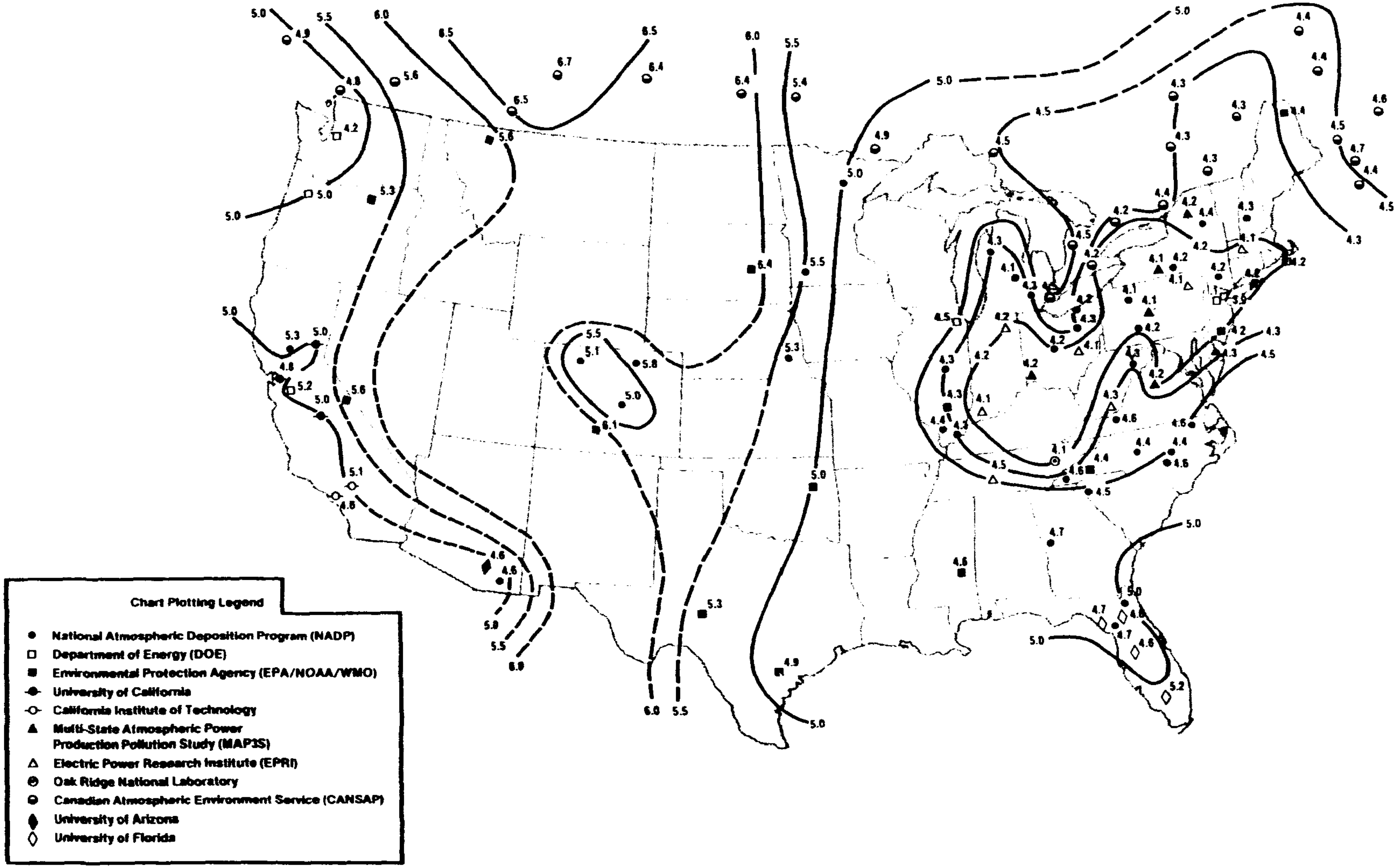


Figure 7. Weighted mean pH of precipitation in the continental United States (1976-1979). (From Wisniewski and Keitz 1982).

high-temperature combustion of fossil fuels. Table 8 presents estimated annual emissions of sulfur and nitrogen oxides, by source, for the nation while Table 9 depicts the distribution of these emissions by EPA regions. Estimated trends in sulfur dioxide emissions through the year 2000 are given by source in Figure 8; similar trends in nitrogen oxide emissions were shown in Figure 2.

Table 8. Estimated national anthropogenic emissions of acid rain precursors in 1977.

Pollutant Source	Annual Production (10^6 metric tons/year)	
	SO_x	NO_x
Transportation	0.8	9.2
Fuel Combustion (Stationary Sources)	22.4	13.0
Industrial Processes	4.2	0.7
Solid Waste Disposal	0	0.1
Miscellaneous	0	0.1
Totals	27.4	23.1

(From USEPA 1978b)

Primary sulfate and sulfuric acid emissions are also released during fuel combustion, augmenting ambient levels of the secondary sulfates. For a given sulfur content of fuel, the combustion of oil yields 5 to 10 times more primary sulfate per unit of energy than coal burning, presumably due to catalytic oxidation of SO_2 by the excessive vanadium content of fuel oil (Homolya and Fortune 1978). Moreover, flue-gas concentrations of H_2SO_4 can be 3-8 times higher in an oil-fired boiler than a coal-fired unit. Oil burning alone is responsible for 97 percent of all primary sulfate emissions associated with combustion processes (Nader 1980).

Table 9. Regional emissions of sulfur and nitrogen oxides compared to population (percent of U.S. totals).

EPA region	States	% of U.S. population	% of U.S. SO_x total	% of U.S. NO_x total
I	CT, ME, MA, NH, RI, VT	5.6	2.1	3.4
II	NJ, NY, PR, VI	12.9	5.3	7.0
III	DE, DC, MD, PA, VA, WV	11.1	15.0	10.7
IV	AL, FL, GA, KY, MS, NC, SC, TN	16.3	21.5	17.5
V	IL, IN, MN, MI, OI, WI	20.6	29.0	23.0
VI	AR, LA, NM, OK, TX	10.3	9.0	17.0
VII	IA, KS, MO, NE	5.3	6.6	6.5
VIII	CO, MT, ND, SD, VT, WY	2.9	2.9	3.7
IX	AZ, CA, HI, NV, GU, AS	11.7	7.3	8.1
X	AK, ID, OR, WA	3.3	1.3	3.1

(From GCA Corporation 1981)

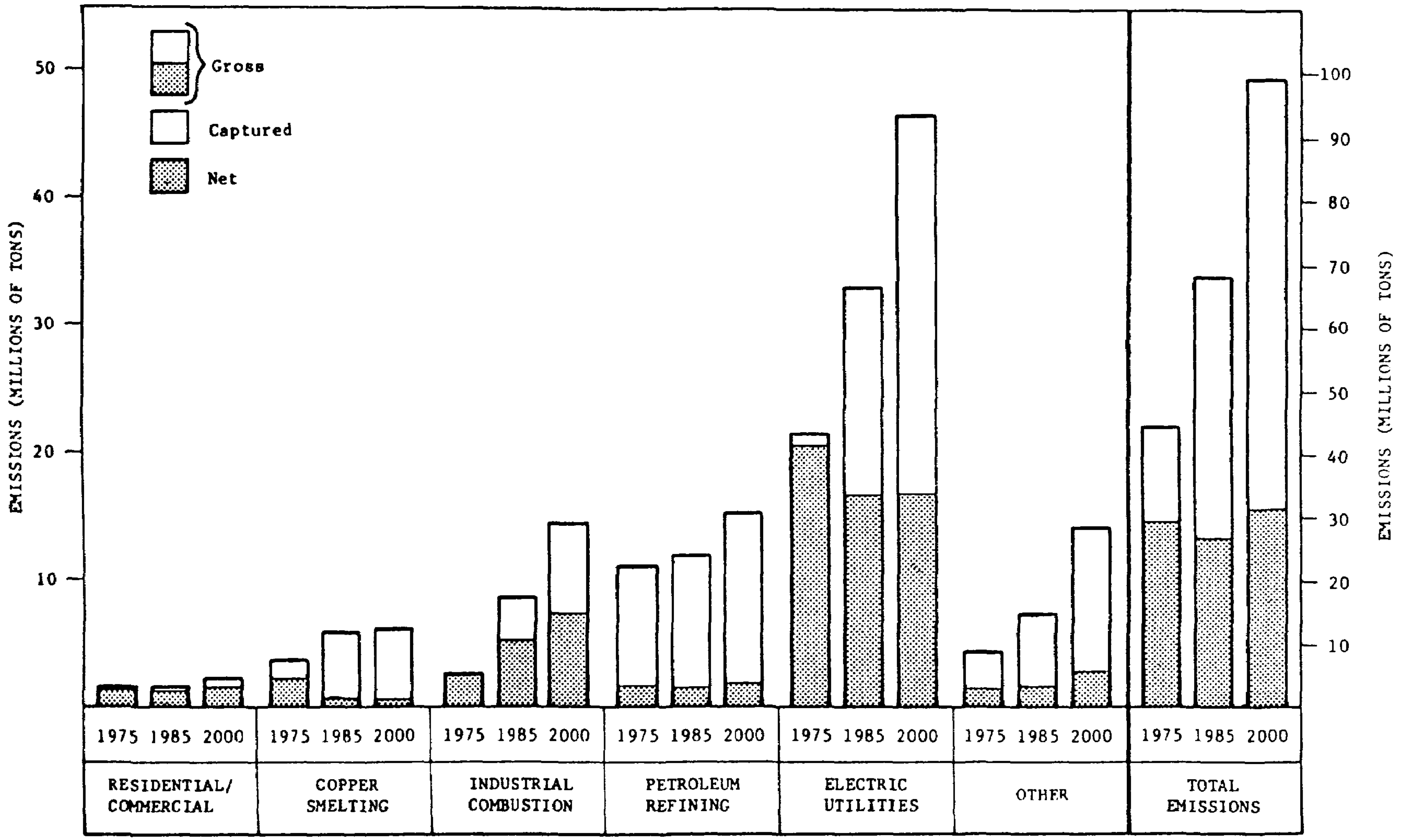


Figure 8. Estimated trends in anthropogenic sulfur oxide emissions by source (high growth scenario). (From USEPA 1980b).

Natural sources of SO_x include the oxidation of biogenic hydrogen sulfide (H_2S), dimethyl sulfide, and other reduced volatile compounds of sulfur, as well as the bacterial reduction of sulfate (Galloway and Whelpdale 1980). Such emissions can be expected where anaerobic conditions exist in swamps, marshes, bogs, and tidal flats. Gaseous flux of biogenic sulfur have been quantified for a variety of these sources and for different soil orders of the United States (Hitchcock 1976; Adams et al. 1980; Aneja 1980). Volcanoes and forest fires sporadically release large amounts of sulfur oxides. Near oceans, sulfates in sea spray are an additional source of atmospheric sulfur.

Globally, biogenic and other natural emissions exceed anthropogenic emissions by a factor of two; man-made emissions are nevertheless projected to equal or exceed naturally-derived atmospheric sulfur on a global basis by the year 2000 (Kellogg et al. 1972; Cullis and Hirschler 1980). On a regional scale, natural sources are reported to currently account for about 4 percent of total sulfur emissions in eastern North America (Galloway and Whelpdale 1980).

Sulfur dioxide and sulfate concentrations are greatest in urban and industrial areas of high emission density. Sulfate levels can vary from in excess of 80 $\mu g/m^3$ in highly polluted localities to concentrations as low as 0.04 ug/m^3 in remote areas (Whitby 1978). Annual sulfate averages of 14-20 ug/m^3 are common in urban areas, while non-urban sites of the northeastern United States average at least 5 ug/m^3 (Altshuller 1976). Sulfate concentrations frequently exceed 15 ug/m^3 in the Ohio River Valley and average 10-15 ug/m^3 over large parts of the eastern United States (Husar et al. 1978). As shown in Figure 9, ambient sulfate levels are generally highest in summer (MacCracken 1979; Altshuller 1980).

Sulfur dioxide concentrations in urban air decreased 67 percent in the period from 1964 to 1970; a 7 percent decline in SO_2 emissions between 1970 and 1979 accompanied a 44 percent drop in national average SO_2 levels (USEPA 1980a). Improved ambient air quality has resulted from restrictions of the sulfur content of fuels, better controls on existing sources, dispersal of sources from urban areas, and the building of taller stacks. Nevertheless, the proliferation of point sources in rural and remote areas has prevented the widespread attainment of federal SO_2 standards (USEPA 1980a). Emissions are remaining at high levels (Glass et al. 1978; Glass 1979b) and are projected to increase both nationally (Altshuller and McBean 1980; USEPA 1980b) and globally (Barnes 1979). Furthermore, reported declines in SO_2 levels have been accompanied by only modest decreases in concentrations of ambient sulfates (Altshuller 1980).

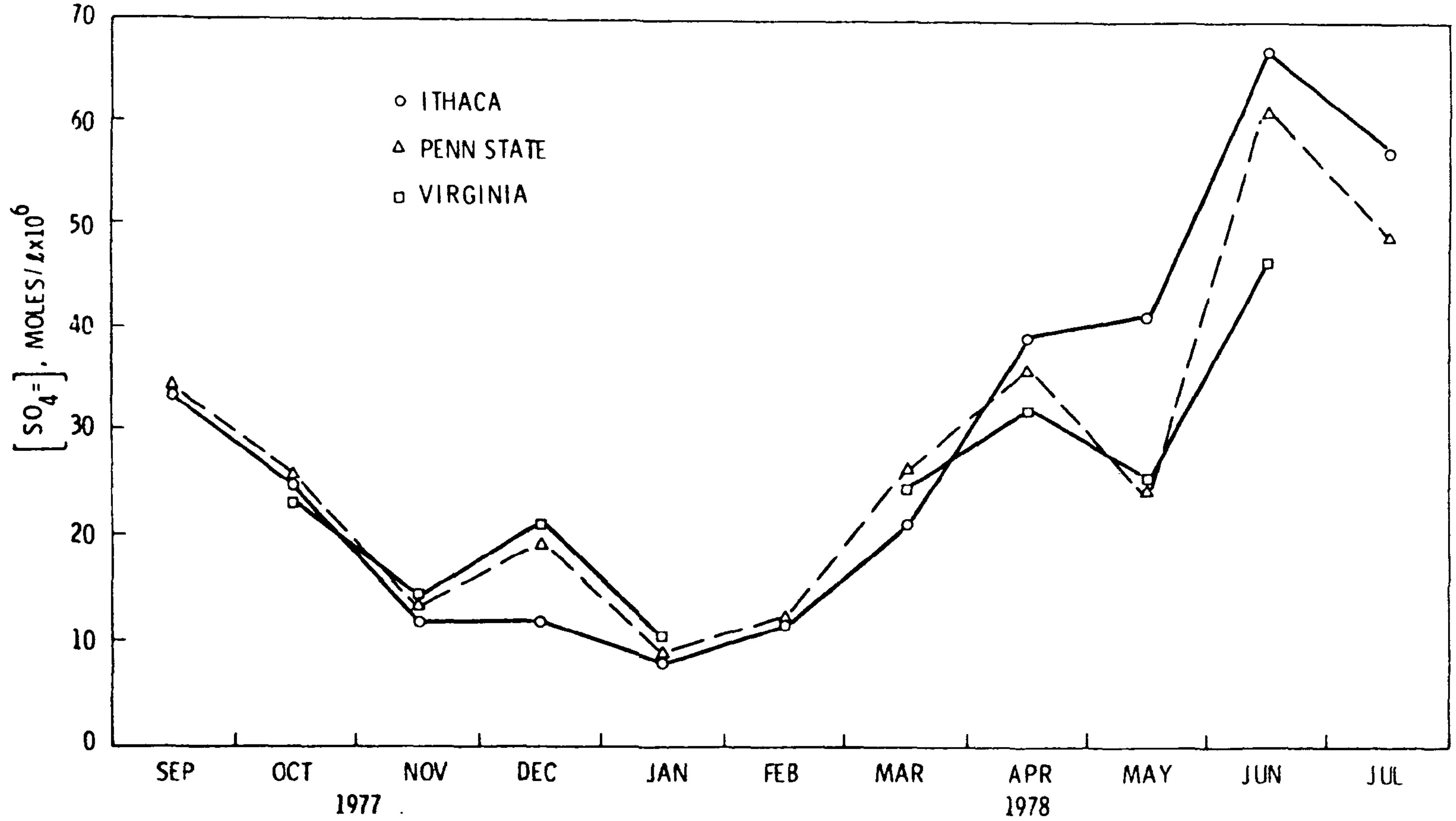


Figure 9. Monthly mean concentrations of sulfate as a function of time at Cornell University (Ithaca), Pennsylvania State University and the University of Virginia. (From MacCracken 1979).

3.0 ECOSYSTEM EXPOSURE TO AIR POLLUTION

Ambient concentrations of air pollution are generally considered to be a function of rates of emission, dispersion, transport, transformation, and deposition. In the past, ecosystem exposures via these processes were only thought to occur in the immediate vicinity of point and area sources. Air pollution, in the form or urban or industrial plumes, was regarded strictly as a discrete, localized phenomenon causing frequent violations of air quality standards at ground level. Efforts to improve local air quality resulted in more efficient pollutant dispersion, longer transport trajectories, and an increased residence time during which chemical transformation of anthropogenic pollution can take place. Evidence of improved ground-level air quality led to general assumptions that ecosystem exposures were on the decline.

Ambient air concentrations continue to be determined by these same fundamental processes, however, in the face of increased pollutant dispersion, the range and complexity of these processes has greatly expanded and they now take place on a regional, and even intercontinental scale. Along with growing recognition of the importance of LRTAP, and the complex integration of air pollutants within global biogeochemical cycling, has come the understanding that injurious substances may be deposited over vast regions, often entering remote ecosystems in quantities equal to or exceeding those deposited near sources. This understanding has led to widespread concern that living resources and their supporting habitats are subject to impact on a scale never before anticipated. This chapter describes in very general terms the atmospheric transport, transformation, and deposition processes that lead to widespread ecosystem exposures to air pollution. It also addresses the different pathways by which living ecosystem components (i.e., fish, wildlife, and their habitats) are exposed to deposited air pollutants.

3.1 ATMOSPHERIC TRANSPORT AND TRANSFORMATION

Atmospheric movements affecting pollutant mixing and transport range from small gusts and eddies, a few meters wide and of minimal duration, to wind systems that may extend over thousands of kilometers and last for 24 hours or more (Bolin et al. 1971). The long-range transport of air pollutants reduces specific pollutant concentrations through mixing and deposition processes while allowing sufficient time for the transformation of primary pollutants to secondary ones. The distance a pollutant travels and potential transformations which can occur depend in large part on site-specific factors such as microclimate and topography, as well as the residence time of the pollutant in the atmosphere, the presence of catalysts, and other factors affecting the efficiency of photochemical reactions.

The formation of photochemical oxidants is a cyclical process, recurring daily in regions where there is inadequate air movement and dispersion of primary pollutants. The reactions producing the oxidants involve the oxidation of hydrocarbons in the presence of nitrogen oxides, sunlight, and several oxidizing agents, of which the hydroxyl radical (OH·) is the most important. Although extremely complex, the chemical reactions that take place can be summarized as follows (Finlayson and Pitts 1976):

$$NO_x + HC \xrightarrow{UV} O_3 + PAN + H_2O_2 + \ldots$$

where

HC = hydrocarbon

NO_x = oxides of nitrogen

O_3 = ozone

PAN = peroxyacetylnitrate

UV = solar ultraviolet radiation providing the energy required for the reaction

These substances are subject to short-range (urban-scale), intermediate-range (mesoscale), and long-range (synoptic scale) transport, during which ozone production per unit of precursor may actually be enhanced (USEPA 1978a). More detailed information on the chemistry involved in photochemical oxidant formation is provided by Altshuller and Bufalini (1971), the National Research Council (1977), and the U.S. EPA (1978a).

The atmospheric reactivity of particulate matter is as diverse as its chemical composition. Moreover, a significant portion of ambient concentrations, called secondary particulate matter, is formed by the conversion of gaseous pollutants, already present in the atmosphere, to liquid aerosols or solid particles. For example, a variety of secondary trace organic compounds are formed by photochemical reactions involving the primary hydrocarbons. Over twenty different organic micropollutants, some of them known carcinogens, have been identified in polluted air masses from western Europe (Lunde and Bjorseth 1977; Lunde et al. 1977). Most of these gas-to-particle conversion processes are dependent on the presence and intensity of solar radiation, the relative humidity and the concentration of oxidizing substances in the air (National Atmospheric Deposition Program 1978).

The predominant reactants and physico-chemical processes involved in particulate transformations are depicted in Figure 10. These processes are related to particle size in order to indicate likely deposition processes and the potential for long-range transport. The coarse particles, along with materials adsorbed to them, are quickly deposited by gravitational settling while the finer fractions may be carried considerable distances from their emission sources. Some of the fine particles, notably

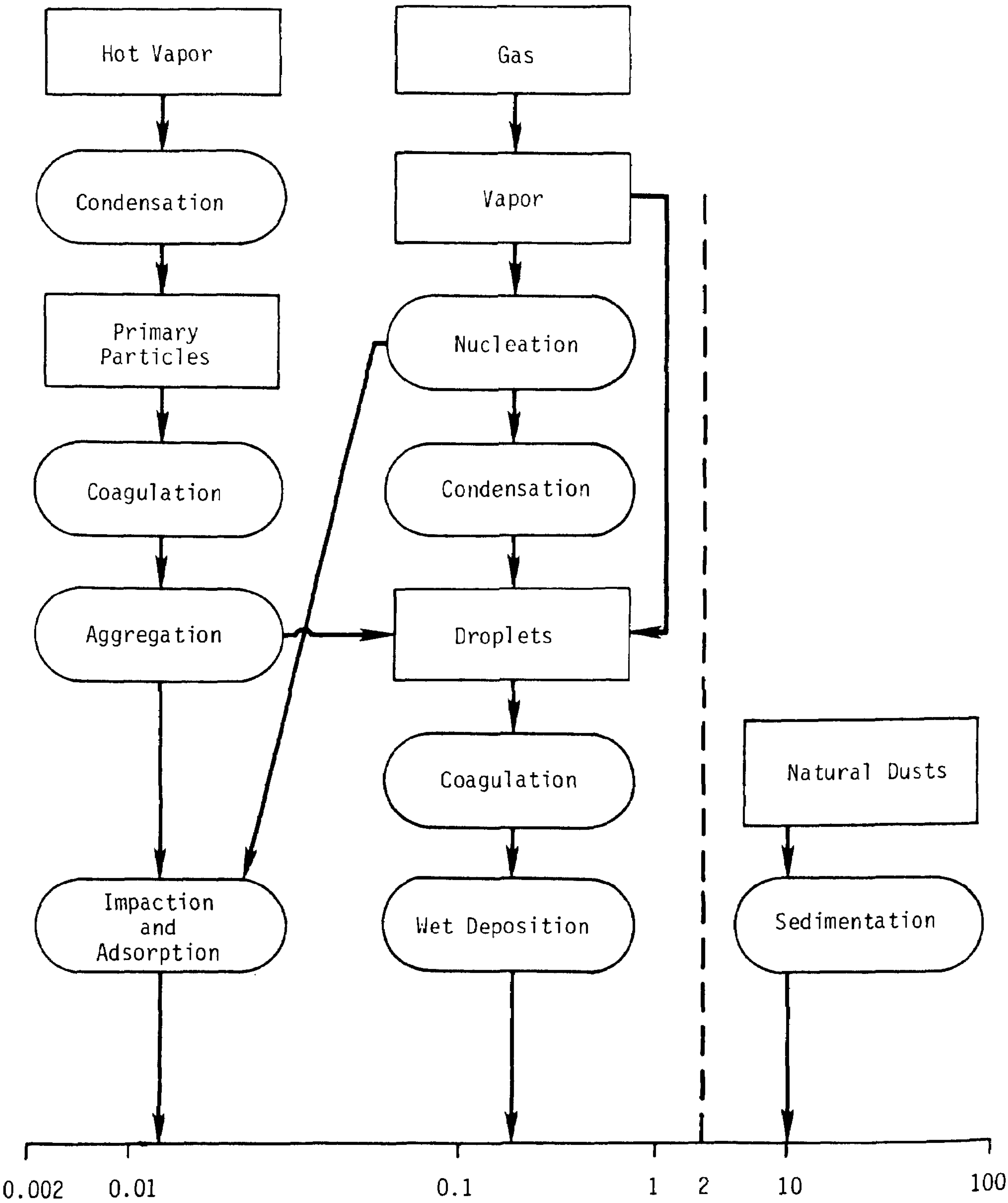


Figure 10. Particulate transformation and deposition processes in relation to particle size. (Adapted from Whitby 1978).

sulfates and nitrates, are hygroscopic in nature: they possess the ability to accelerate the condensation of water vapor. This property can simultaneously affect their size, shape, reactivity, and pH. An excellent review of particulate movement and reactivity in the atmosphere has been prepared by Whitby (1978).

The primary acidifying gases, oxides of sulfur and nitrogen, can also be transported great distances. While airborne they are transformed in the atmosphere to secondary acidifying particulates. These aerosols may be transported hundreds of kilometers further than the primary pollutants since they deposit at much slower rates (Barnes 1979).

In clean air, SO_2 oxidizes slowly to form sulfur trioxide (SO_3), which is generally present in only minor amounts in the atmosphere because it reacts rapidly with moisture to form sulfuric acid (H_2SO_4). The reactions producing sulfuric acid can be summarized as follows (Vermeulen 1978):

$$SO_2 \rightarrow \left\{ \begin{array}{l} 2\ SO_2 + O_2 \longrightarrow 2\ SO_3 \\ \\ SO_3 + H_2O \longrightarrow 2\ H^+ + SO_4^{=} \end{array} \right\} \rightarrow H_2SO_4$$

These reactions may be effectuated through (Eggleton and Cox 1978; Dovland and Semb 1980; McMurray 1980):

- homogeneous oxidation processes, or the photochemical oxidation of SO_2 gas by thermally produced reactants or photochemically generated free radicals (e.g., $OH\cdot$, $\cdot HO_2$); and

- heterogeneous oxidation processes, or the catalytic oxidation of SO_2 adsorbed to aqueous aerosols by metal ions (e.g., iron, manganese) in solution.

Both of these conversion processes produce aerosols in the sub-micron size range.

Atmospheric nitrates result from similar reactions which oxidize nitrogen oxide (NO) and nitrogen dioxide (NO_2) to nitric acid (HNO_3) and other inorganic and organic nitrates. The following general mechanism for the conversion of NO_x to nitric acid has been postulated (Vermeulen 1978):

$$SO_2 \rightarrow \left\{ \begin{array}{l} 2\ NO + O_2 \longrightarrow 2\ NO_2 \\ 3\ NO_2 + H_2O \longrightarrow 2\ H^+ + 2\ NO_3^- + NO \\ 4\ NO_2 + 2\ H_2O + O_2 \longrightarrow 4\ H^+ + 4\ NO_3^- \end{array} \right\} \longrightarrow HNO_3$$

In addition, nitrate salts may be formed in the atmosphere through a variety of reactions. The direct homogeneous capture of gaseous nitric acid by gaseous ammonia:

$$NH_3 + HNO_3 \longrightarrow NH_4NO_3$$

may be a significant source of ammonium nitrate salt in the atmosphere if ammonia levels in the polluted atmosphere are sufficiently high (National Research Council 1977). Nitric acid also forms some mixed compounds or complexes of considerable stability in sulfuric acid solutions, and may become incorporated within sulfuric acid droplets at the lower temperatures of the upper atmosphere. In general, much less is known of exact mechanisms of nitrate formation as most research has focused specifically on processes of atmospheric sulfate formation.

Considerable research has been undertaken to quantify rates of sulfur dioxide conversion to sulfate in ambient air, with widely varying results (MacCracken 1979). Ratios of sulfur dioxide to sulfate in urban and industrial plumes have been suggested as indicators of SO_2 transformation rates; these ratios are greatest near emission sources and may serve as a surrogate measure of the age of plumes (Dovland and Semb 1980). Homogeneous oxidation of SO_2 to $SO_4^=$ can occur at rates up to 1.0 percent per hour in clean air during summer and may reach values of 5 percent per hour in irradiated urban air (Eggleton and Cox 1978). Homogeneous oxidation in plumes of coal-fired power plants typically occurs at rates less than 1.0 percent per hour in clean air, however diurnal variations also occur and maximum rates of 3 percent per hour have been recorded at midday (Newman, L. 1980). Conversion rates for heterogeneous oxidation are similar, averaging 1 to 2 percent in unpolluted air and 2 to 6 percent in urban atmospheres (Dovland and Semb 1980). However, known reactions alone are insufficient to explain SO_2 conversion to $SO_4^=$ in the patterns and quantities observed, suggesting that one or several unknown mechanisms may play a significant role in secondary pollutant formation (Budiansky 1980).

Comparisons of SO_2 emissions with ambient sulfate concentrations in specific regions indicate that sulfate distribution is determined primarily by transport and transformation processes and not local sources of SO_2 (Altshuller 1976, 1980). Air parcel trajectory analysis has been employed to determine the origins of elevated sulfate concentrations in ambient air. The long-range transport of air pollutants has been found to be greatest in air masses that have stagnated for several days over heavily industrialized regions. In New York, elevated sulfate concentrations and precipitation acidity have been correlated primarily with westerly and southwesterly transport from Canada and the Ohio River Valley (Galvin et al. 1978), while in Scandinavia they have been associated with southerly and southwesterly transport from industrial centers of western Europe and the United Kingdom (Rodhe et al. 1972).

3.2 ATMOSPHERIC DEPOSITION PROCESSES

The removal of primary and secondary pollutants from the atmosphere occurs through complex physical and chemical processes referred to as atmospheric deposition, which may be defined as the transfer from air to ground of any gas or particle via wet and dry removal processes; acid deposition refers to transfers of strictly acidic substances by these same removal processes (Interagency Task Force on Acid Precipitation 1981).

Wet deposition is defined as the amount of material removed from the atmosphere by rain, snow, or other precipitation forms; the term is also used to refer to the process by which gases, liquids, or solids are transferred to the ground during a precipitation event (Interagency Task Force on Acid Precipitation 1981). Two separate removal processes are involved (Hales 1972; Fowler 1980a, b):

- rainout (or snowout), in which pollutants serve as condensation nuclei or are incorporated into hydrometeors, or water vapor condensates, before they begin to fall; and

- washout, the incorporation of pollutants into falling hydrometeors below the cloud.

Collectively, these processes are referred to as precipitation scavenging (Slinn 1977).

The concentration of pollutants within droplets depends on the initial concentration in the collector particles, pollutant concentrations in droplets added to the initial particle, and amounts lost prior to impact (Hales 1972). The time available for mixing in contaminated atmospheres and the rate at which vapor droplets are scavenged from the cloud by larger raindrops both affect the volume of pollutants deposited with precipitation.

As precipitation nears the earth's surface, it may be characterized in several ways (Eaton _et al_. 1973; Galloway and Parker 1980):

- incident wet deposition is the rainfall transferred directly to the ground by gravitational forces;

- throughfall is rain that has passed through a leaf canopy;

- net throughfall is the chemical composition of water that has passed through a leaf canopy (i.e., the sum of materials in incident wet deposition plus substances leached or washed from leaves minus materials retained within the leaf canopy); and

- stemflow is rainwater which flows down the branches and trunks of vegetation.

The special event of fog is another wet removal process which may be significant in the deposition of air pollutants. Fogs are known to be excellent scavengers of acid aerosols (Smith et al. 1980). Studies have shown that the acidity of cloud water often exceeds that of precipitation, suggesting that frequent fog, for example in mountainous areas, may be a significant source of acidic inputs (MacCracken 1979).

Wet deposition processes also remove metal and trace elements from the atmosphere. Exceptions are the vapor-phase metals and organics for which gaseous adsorption is the dominant removal mechanism (Galloway et al. 1981). Table 10 quantifies average concentrations of trace metals in wet deposition from remote, rural, and urban areas. The ratios provided in Table 10 indicate concentration factors by which urban and rural quantities exceed those in remote areas.

Concentrations of sulfuric and nitric acid in precipitation have been monitored during the past several years by a number of national and regional precipitation chemistry networks. Patterns have emerged which permit the characterization of trends in acid precipitation for all regions of the continental United States. Weighted mean pH values of precipitation across the United States for the period 1976-1979 were depicted in Figure 7. Mean annual precipitation patterns, when combined with these pH determinations, permit the calculation of mean annual hydrogen ion deposition in precipitation, as shown in Figure 11.

Dry deposition is defined as the aggregate of all materials transferred from the atmosphere to natural surfaces in the absence of precipitation; its definition also includes the physical processes of transfer (Interagency Task Force on Acid Precipitation 1981). Dry removal processes include (Fowler 1980a, b; Galloway and Parker 1980):

- dry fallout (or sedimentation), the gravitational settling of particles greater than 10 um in diameter;
- aerosol impaction, the wind driven deposition of sub-micron materials; and
- gaseous adsorption, the natural attraction between gases and solid or liquid surfaces.

Because it is easy to collect precipitation, the wet deposition of atmospheric pollutants has generally been easier to characterize on a qualitative and quantitative basis than dry deposition, although many questions remain about the exact nature of both processes (Galloway et al. 1979; Galloway and Parker 1980). Rates of dry deposition are virtually impossible to predict with any degree of assurance since they are controlled by numerous factors including some related to the pollutant, such as size, shape, and chemical properties, as well as meteorological conditions (McMahon et al. 1976; Sehmel 1980). Dry deposition rates cannot be measured with certainty because the amount of material transferred will also depend on the nature of the receiving surface.

Table 10. Average concentrations of metals in wet deposition. Ratios at remote, rural, and urban sites provided are factors of concentration of urban and rural to remote levels.

Metal	Concentrations (ug/l)			Ratios	
	Remote	Rural	Urban	Urban/Remote	Rural/Remote
Ag	0.008	0.023	3.2	400	3
As	0.019	0.04	-	-	2
Cd	0.008	0.6	0.7	87	75
Co	-	0.01	1.8	-	-
Cr	-	0.27	3.6	-	-
Cu	0.055	5.3	30	554	96
Hg	0.048	0.11	1.0	21	2
Mn	0.22	10.0	25	113	45
Mo	-	-	0.2	-	-
Ni	0.1	1.5	17	170	15
Pb	0.14	15	41	292	110
Sb	0.034	-	-	-	-
V	0.022	1.1	68	3,090	50
Zn	0.22	45	40	181	200

(From Galloway et al. 1981)

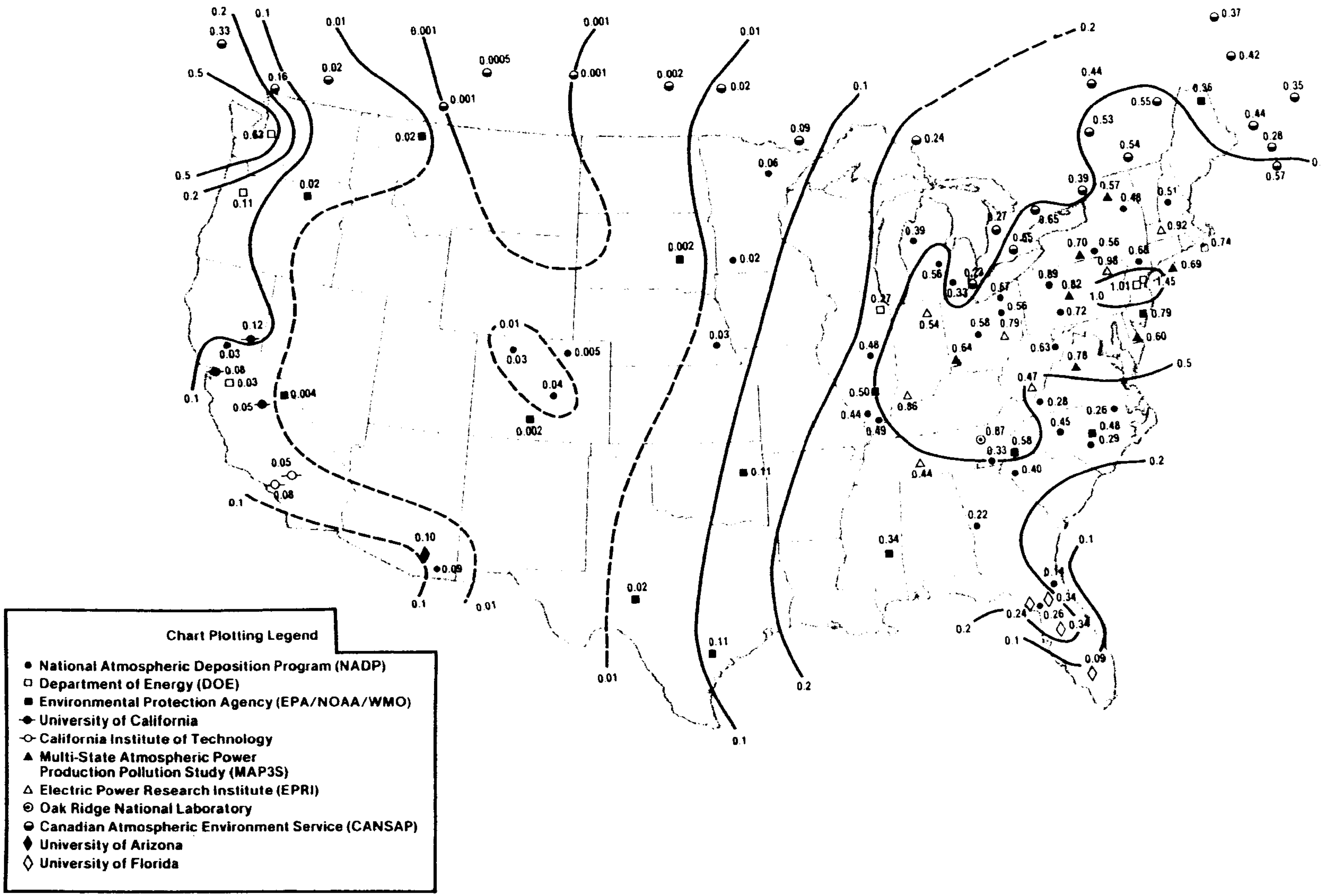


Figure 11. Mean annual hydrogen ion (H^+) deposition in precipitation over the continental United States, 1976-1979 (Kg/ha). (From Wisniewski and Keitz 1982)

Despite the high degree of uncertainty in the understanding of dry deposition processes, it is known that they are quite important. Adsorption is the most important removal process for gaseous pollutants, including SO_x, NO_x, and HNO_3. Particulate matter is deposited by impaction and gravitational settling. In arid lands, as would be expected, dry deposition processes are the dominant mechanism for pollutant deposition, but in non-arid lands as well, the amount of matter deposited via dry removal processes may exceed the amount deposited by wet removal processes (Gravenhorst et al. 1980).

Special events of dew and frost must be considered in connection with dry depositon processes because of their potential for increasing the acidity of deposited pollutants. Dews, for example, dissolve previous gaseous and particulate deposits on plant surfaces and enhance their transformation to strong acids, leading to a rapid decrease in the pH of the wetted plant surfaces. The contribution of frost to acidity is only significant when it melts and assumes the characteristics of dew because ice does not usually incorporate foreign substancesinto its crystalline structure (Smith et al. 1980).

3.3 PATHWAYS OF AIR POLLUTION EXPOSURE TO FISH, WILDLIFE, AND THEIR HABITATS

Atmospheric contaminants enter the different compartments of aquatic and terrestrial ecosystems through a variety of pathways. The degree of exposure depends on pollutant emission rates, proximity to emission sources, characteristics of pollutant transport, transformation and deposition, as well as modes of pollutant uptake and distribution within biotic or abiotic compartments. This section discusses pathways by which fish, wildlife and living components of their habitats are physically contacted by atmospheric pollutants.

The most direct exposures of terrestrial wildlife to air pollution occur through the inhalation of gases and particulates. Fine particles (< 1.0 um in diameter), like gases, bypass respiratory filters and can be deposited deep within animal lungs (Davidson et al. 1974; Natusch et al. 1974). Airborne particulates and reactive gases, such as ozone and sulfur dioxide, may adsorb to the eyes of animals (Newman 1980). Direct exposures of terrestrial vertebrates to acid precipitation are of little consequence due to the dilute nature of dissolved acids and the protection afforded body surfaces by scales, feathers, or fur.

Terrestrial wildlife may be indirectly exposed to air pollutants by ingesting contaminated vegetation. Examples of such exposures include the ingestion of plants with fluorides or arsenic deposited on their surfaces (Lerman and Darley 1975). Animals may also ingest air pollutants with drinking water. Similarly, indirect exposures can occur from the ingestion of food organisms that have accumulated various air pollutants (Stickel 1975).

Terrestrial plants comprising wildlife habitats are exposed to atmospheric pollution through contact with above- and below-ground plant parts. Exposure pathways depend on whether the pollutant is in gaseous or particulate form. Gaseous air pollutants contact internal leaf tissues after entry through leaf stomata. This exposure mechanism has been verified for sulfur dioxide (Mudd 1975a), nitrogen oxides (Taylor et al. 1975), and ozone (Heath 1975). Plant exposure to peroxyacetylnitrate has not been thoroughly studied, however passage through leaf stomata appears important (Mudd 1975b). Soils may act as a sink for gaseous pollutants which are adsorbed by microorganisms or taken up by plants.

Aerial deposition is the primary mechanism of plant exposure to particulates (Lerman and Darley 1975). Fine particles, including trace elements, fluorides, and acid aerosols, are able to penetrate leaf tissues via opened stomata (Chang 1975). Coarse particulates collect on leaf surfaces and may form hardened crusts through interactions with moisture on leaf and stem surfaces. Acidic substances dry-deposited on leaves can be dissolved by dew or frost (Smith et al. 1980), and these, along with dilute acids in precipitation, collect in leaf depressions and enter via epithelial cracks near specialized structures (e.g., stomata, veins, and trichomes) of the leaf surface (Evans 1980).

Soil microorganisms may be exposed to particulate matter deposited on soils; these substances may also be taken up by plant roots. Field studies have shown that approximately 90 percent of deposited metals are retained in the top 15 centimeters of soils (Buchauer 1973). There they may be readily absorbed by roots. Plants absorb and accumulate both essential and non-essential trace elements from dilute soil solutions. Root exposures may be further augmented by soil acidification and other pollutant-induced alterations that increase the biological availability of trace contaminants (Zimdahl 1976). Aerial portions of plants may ultimately be affected by pollutants absorbed through the roots if the pollutants are readily translocated within the plant.

Like plant roots and soil microbes, freshwater fauna may be exposed to relatively unchanged forms of air pollutants, their transformation products and substances mobilized by pollution effects. For example, an increased availability of toxic metals will result directly from wet and dry deposition as well as indirectly through increased releases from sediments and the watershed caused by acid precipitation (Beamish and Van Loon 1977). Trace elements found to increase in acidified waters include aluminum, cadmium, iron, magnesium, and manganese (Hall and Likens 1980; Hall et al. 1980).

Aquatic organisms are exposed to these same types of pollutants primarily through adsorption and uptake across gills and other permeable membranes. This mechanism is particularly important for hydrogen ions, toxic metals (e.g., aluminum and mercury), and the lipophilic organic substances (Muniz and Lievestad 1980a). Other exposures to air pollutants occur through the ingestion of contaminated food, water, detritus or sediment.

Elevated exposures can be expected to coincide with spring snowmelt or intense precipitation events; the acidity associated with such events increases the abundance of biologically available forms of metals (Harvey 1979; Troutman and Peters 1980). Aerial exposures to ambient air pollution may be important for those organisms which inhabit or feed at the water's surface.

Air pollutants contact emergent aquatic vegetation by direct deposition on plant surfaces or by entry through stomata. Submergent and floating forms may adsorb contaminants in the water column to exterior surfaces and transport them across membranes. For example, trace metals may be adsorbed to cell walls and transported through them to internal plant tissues (Dvorak et al. 1978). Rooted forms may also absorb and translocate atmospheric contaminants trapped in sediments.

Exposure pathways are key determinants of the nature of biotic responses to air pollutants. Depending on its mode of entry and site of deposition, a given pollutant may affect an organism in different ways. Therefore, assessments of the biological impacts of air pollution and acid rain should commence with a determination of the relative importance of various exposure pathways among the organisms to be investigated.

4.0 FACTORS AFFECTING ECOSYSTEM SENSITIVITY

Many interacting variables determine the vulnerability of aquatic and terrestrial ecosystems to air pollution and acid rain phenomena. They are of two basic types:

- factors related to the intensity and distribution of pollutant loadings to ecosystems; and
- factors related to the inherent susceptibility of ecosystem structure and function to alterations induced by atmospheric deposition.

Ecosystem impact is usually, but not always, associated with the occurrence of both types of factors. Inherently susceptible ecosystems, for example, may be defined as sensitive even in the absence of pollutant deposition because they would be subject to alteration in the event that airborne loadings increase.

The primary factors regulating ecosystem sensitivity are (National Atmospheric Deposition Program 1978; USEPA 1980c):

- meteorology;
- geology;
- pedology;
- hydrology;
- hydrochemistry;
- topography;
- biota; and
- human activity.

Each of these factors influences the extent of pollutant loadings or ecosystem responses. Together, they may render certain ecosystems much more vulnerable to adverse impacts than others. In general, the presence or absence of these factors largely determines whether or not biological effects will occur; the complexity of their interactions moderates biotic effects in such a way that consistent patterns are difficult to establish (Shriner 1979).

4.1 METEOROLOGY

Meteorological factors contributing to ecosystem sensitivity are both complex and varied. In all ecosystems, the dilution and dispersion of primary pollutants are regulated by convection, turbulence and wind patterns. Other influences include atmospheric stability and the frequency of periodic thermal inversions. These factors determine the residence time of pollutants in the atmosphere which in turn affects their distribution and long-range transport as well as the probability of their conversion into secondary pollutants.

Humidity and solar radiation levels have a strong influence on the rate of conversion of atmospheric gases to acids. At the time of deposition, wind and humidity together affect rates of gaseous adsorption and particle impaction on terrestrial surfaces. Precipitation amounts and frequencies, as well as pH and chemistry, determine the extent of ecosystem pollution through rainout and washout. Seasonal variations in dry deposition, rainfall, snowfall, and snowmelt can greatly influence the magnitude of atmospheric pollutant inputs. Special events of dew, frost and fog can also significantly augment pollutant loadings in areas of frequent occurrence (Smith et al. 1980). Moreover, snow, rain and special events possess differing efficiencies in the removal and concentration of airborne acids and other pollutants.

In all terrestrial ecosystems, the ratio of precipitation to evapotranspiration influences the extent of mineral release from soils through leaching (National Atmospheric Deposition Program 1978). In the eastern United States, where precipitation generally exceeds water losses to the atmosphere, surface runoff and soil percolation may transport significant amounts of leachates and unbuffered pollutant inputs from terrestrial to aquatic ecosystems.

Several meteorological variables have been demonstrated or hypothesized to mediate the extent of plant injury from ambient air pollution and atmospheric deposition. These include (Evans 1979a; Shriner 1979):

- temperature;
- light intensity;
- relative humidity;
- duration and intensity of precipitation; and
- the time between precipitation events (i.e., the time available for the accumulation of dry deposition).

Meteorological factors also influence diurnal and seasonal variations in the sensitivity of plants to air pollutant loadings (Guderian 1977).

In general, the direction of prevailing winds, the trajectories of polluted air masses and the nature of precipitation patterns exert the greatest influence on the magnitude of ecosystem loadings due to acid deposition. These, along with other meteorological factors discussed above, lead to disproportionately large amounts of atmospheric deposition in mountainous regions, notably the Adirondacks of New York, the Sierras of California, the Rockies of Colorado and the Appalachian range.

4.2 GEOLOGY

The geological characteristics of an ecosystem are generally considered to be the primary determinant of sensitivity to acid deposition (Voigt 1980). Bedrock geology is especially significant where soils are thin and runoff is substantial. In certain localities, however, soils may largely override the influence of geological substrates (Hendrey et al. 1980a).

The extent of ecosystem impact from acid rain is determined by the nature of dominant rock types as these substrates have widely varying capacities to neutralize acid. Granite, quartz, basalt, and other aluminosilicate materials, normally the most resistant to physical and chemical weathering, are least capable of adequately buffering acidic inputs (Abrahamsen et al. 1979; Likens et al. 1979; Hendrey et al. 1980a,b; Norton 1980). The result is an accelerated leaching of nutrient cations, notably sodium, calcium, potassium and magnesium, as well as toxic metals such as mercury and aluminum (Webb 1980). In fact, ecosystem losses of calcium, magnesium and aluminum closely correlate with the amount of hydrogen ions passing through the watershed, and enrichment of drainage waters with these substances may result (National Atmospheric Deposition Program 1978).

A classification of rock types, based on their capacity to buffer acid precipitation, is presented in Table 11. Figures 12 and 13 depict two different methods of mapping geologically sensitive regions utilizing this classification. Type 1 rocks are associated with extensive aquatic ecosystem impact from acid precipitation while Type 2 substrates generally facilitate the acidification of first- and second-order streams and headwater lakes. Type 3 rocks are not thought to contribute to aquatic acidification except where surface run-off drains over frozen ground. Type 4 rocks are considered to have "infinite" buffering capacity and for this reason are in no way associated with aquatic ecosystem effects. Thus, bedrock geology correlates directly with the acidification of waterways and determines the extent of acid rain damage to aquatic ecosystems (Hendrey et al. 1980a; Norton 1980).

Earlier efforts to map geologically sensitive areas of the United States, performed by Galloway and Cowling (1978) and Likens et al. (1979), were based on large scale geological maps (Figure 14). From these and more recent studies, it has become apparent that significant portions of the country are underlain by geologically sensitive bedrock. Those areas of the U.S. underlain by granite bedrock include the Adirondack and

Table 11. Classification of rock types used to distinguish geological sensitivity.

Type 1	Low to no buffering capacity: Granite/syenite, granite gneisses, quartz sandstones, or metamorphic equivalents.
Type 2	Medium to low buffering capacity: Sandstones, shales, conglomerates, high-grade metamorphic felsite to intermediate volcanic rocks, intermediate igneous rocks, calc-silicate gneisses with no free carbonates.
Type 3	Medium to high buffering capacity: Slightly calcareous, low grade intermediate to mafic volcanic rocks, ultramafic and glassy volcanic rocks.
Type 4	High buffering capacity: Highly fossiliferous sediments or metamorphic equivalents, limestones or dolostones.

(From Hendrey et al. 1980a, b; Norton 1980)

Appalachian mountains and large parts of New England, Michigan, Wisconsin, Minnesota, Washington, Idaho, Oregon, California and Colorado (Interagency Task Force on Acid Precipitation 1981). Coastal regions with sand substrates are also known to be vulnerable to aquatic and terrestrial ecosystem acidification (Glass et al. 1980). A large part of eastern New Jersey is reported to have acidified streams and groundwaters where sand substrates are deep (Johnson, A. 1979a, b).

4.3 PEDOLOGY

The ability of soils to neutralize acidification, retain other contaminants and prevent pollutant transfers to aquatic ecosystems depends primarily on soil type, depth, age, parent bedrock, carbonate and organic content, cation exchange capacity, glaciation history and the influences of vegetation. For purposes of identifying vulnerable soils, however, the following parameters are considered to be the most important (McFee 1980a, b; Klopatek et al. 1980):

- soil acidity (pH), which approximates the cation replacement efficiency of additional hydrogen ion inputs;
- cation exchange capacity (CEC), or the total buffering potential provided by the exchange sites of clay and organic matter;

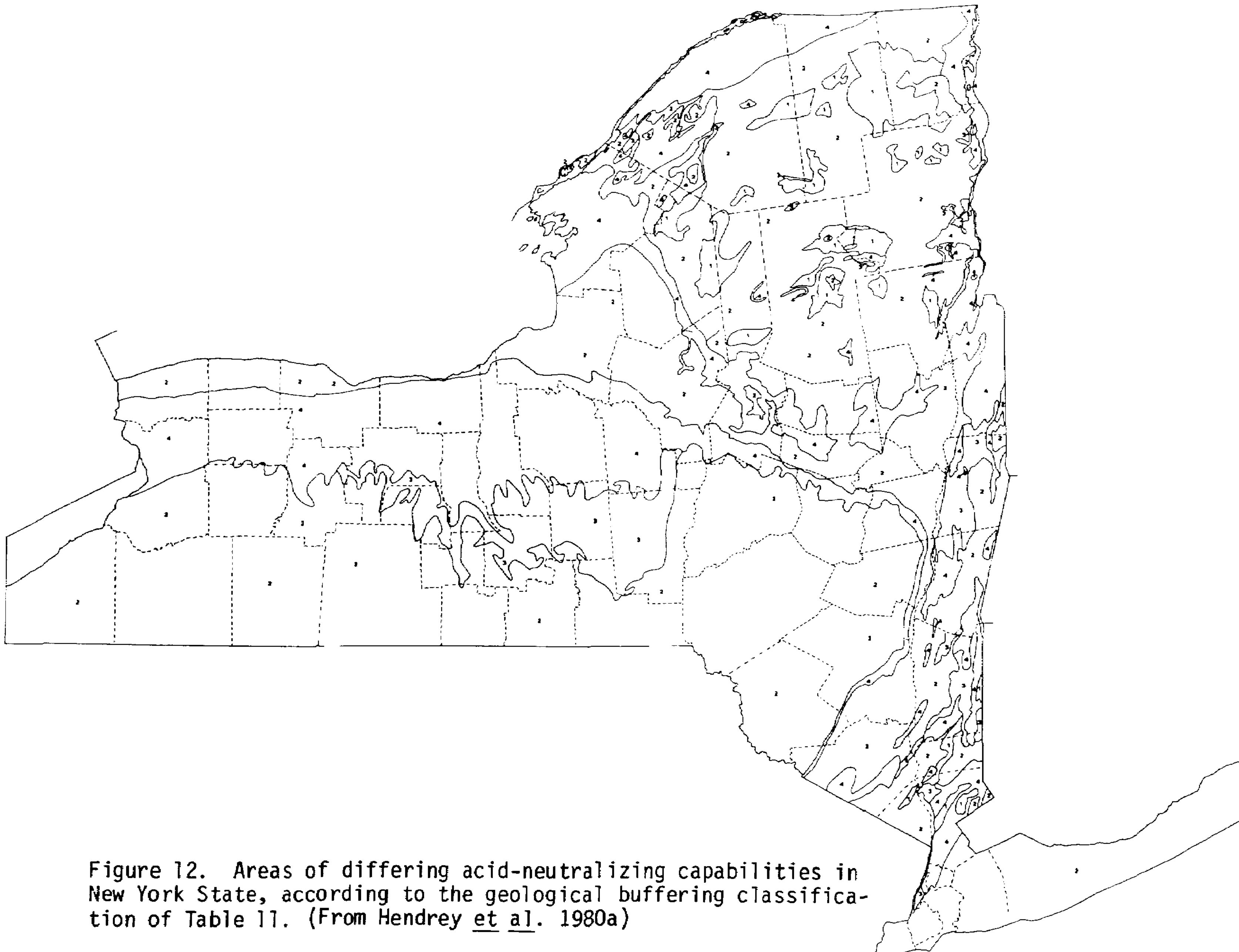


Figure 12. Areas of differing acid-neutralizing capabilities in New York State, according to the geological buffering classification of Table 11. (From Hendrey et al. 1980a)

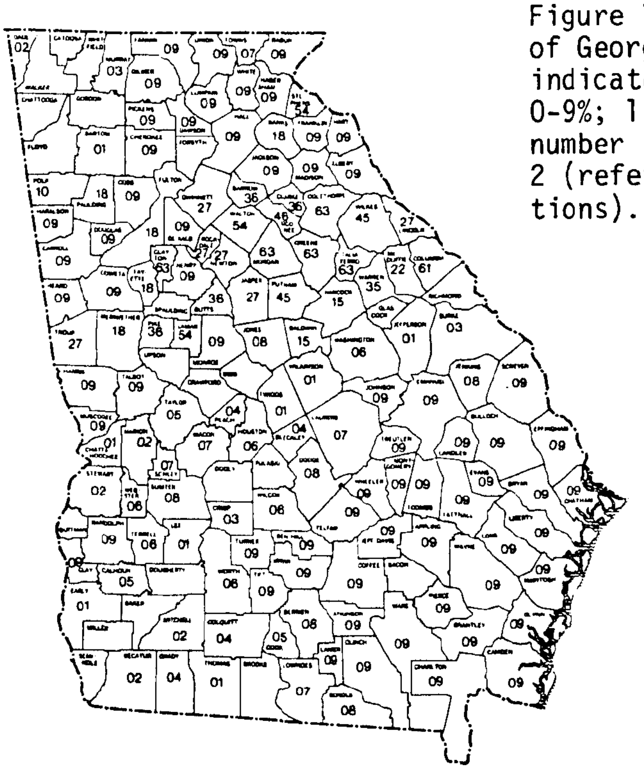


Figure 13. Vulnerability map for the state of Georgia, by county. The first number indicates the percentage of rock type 1 (0, 0-9%; 1, 10-19%; 2, 20-29%; etc.) The second number indicates the percentage of rock type 2 (refer to Table 11 for rock type classifications). (From Hendrey et al. 1980a).

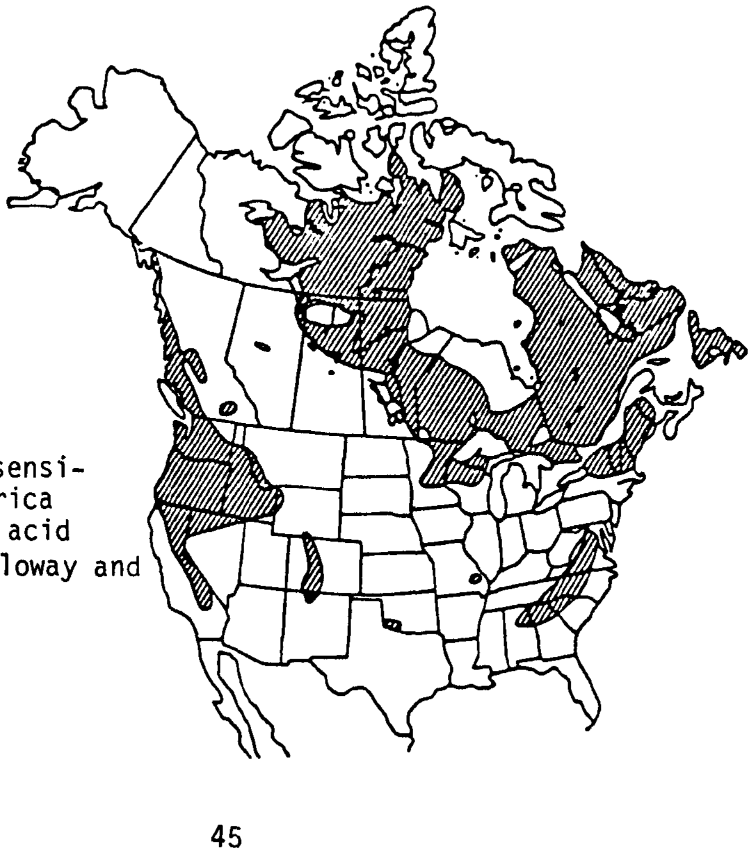


Figure 14. Geologically sensitive regions of North America with lakes susceptible to acid precipitation. (From Galloway and Cowling 1978).

- base saturation of exchange capacity (BS), a measure of the degree of soil development, and a function of pH;

- soil management systems, including cultivation, fertilization and liming, as well as renewal through flooding and other processes; and

- the presence or absence of carbonates within the soil profile.

Vulnerable soils are generally defined as those with potential for decreased productivity in response to acid deposition. They are characterized by low CEC; hence reduced clay and organic content, and fewer sites for proton exchange (McFee 1979, 1980a, b). Sensitive soils possess medium to high base saturation (30-50%) and pH values greater than 5 (Klopatek et al. 1980). This is because soils of low base saturation (podzols) have low leaching rates while soils of high base saturation (loams) have high leaching rates (Abrahamsen 1979). Like susceptible bedrock, sensitive soils are prone to accelerated leaching of nutrient and metal cations. The hydrogen ions in acid precipitation replace other cations in the soil, notably calcium, magnesium, iron and aluminum, which are dissolved in surface run-off and concentrated in the hydrographic network.

A classification of soil sensitivity to acid precipitation is presented in Table 12. Its application to the mapping of sensitive soil regions is exemplified in Figure 15. Noncalcareous, sandy soils of pH 5 are generally agreed to be the most sensitive to acid precipitation (McFee 1980a, b; Wiklander 1980). Mature soils that are naturally acidic are seldom subjected to further acidification from atmospheric deposition, yet they are vulnerable to accelerated leaching, particularly of aluminum, which can have long-term consequences for their sustained productivity (McFee 1980b). These and other soil types are compared in Table 13 on the basis of two major sensitivity parameters. Petersen (1980) provides a detailed discussion of soil sensitivity based on recognized national and international soil classifications.

Remaining soil sensitivity parameters include soil moisture content and the extent of soil-water mixing as water percolates through the soil profile (Voigt 1980). These factors, as well as soil texture, depth and porosity, are particularly important, for if acidic precipitation cannot achieve chemical equilibrium with soils, downstream water bodies will be subject to considerable hydrochemical alteration (Bache 1980). Factors mediating rates of soil acidification include (Wiklander 1980):

- calcium carbonate ($CaCO_3$) content;

- ferro-magnesium mineral content;

- soil texture;

Table 12. Classification of soil sensitivity to acid precipitation based on cation input and the chemical characteristics of the top 25 cm.

Sensitivity Class	Cation Exchange Capacity meq/100g	Other Relevant Conditions
Non-sensitive (NS)	Any value or	Free carbonates present or subject to frequent flooding
	>15.4	None.
Slightly sensitive (SS)	$6.2 \leq CEC \leq 15.4$	Free carbonates absent; not subject to frequent flooding
Sensitive (S)	> 6.2	Free carbonates absent; not subject to frequent flooding

Source: McFee (1980a, b)

Table 13. Soil sensitivity to acid precipitation based on buffering capacity and hydrogen ion retention.

	Calcereous soils	Noncalcereous clays pH > 6	Noncalcareous sandy soils pH > 6	Cultivated soils pH > 5	Acid soils pH > 5
Buffering capacity	Very high	High	Low	High	Moderate
Hydrogen ion retention	Maximal	Great	Great	Great	Slight
Overall sensitivity	None	Moderate	Considerable	None or slight	Slight

(From Wiklander 1979)

Key:

NS - The area contains mostly non-sensitive areas
S1 - Sensitive soils dominate the area
SS1 - Slightly sensitive soils dominate the area
S2 - Sensitive soils are significant, but cover less than 50% of the area
SS2 - Slightly sensitive soils are significant, but cover less than 50% of the area

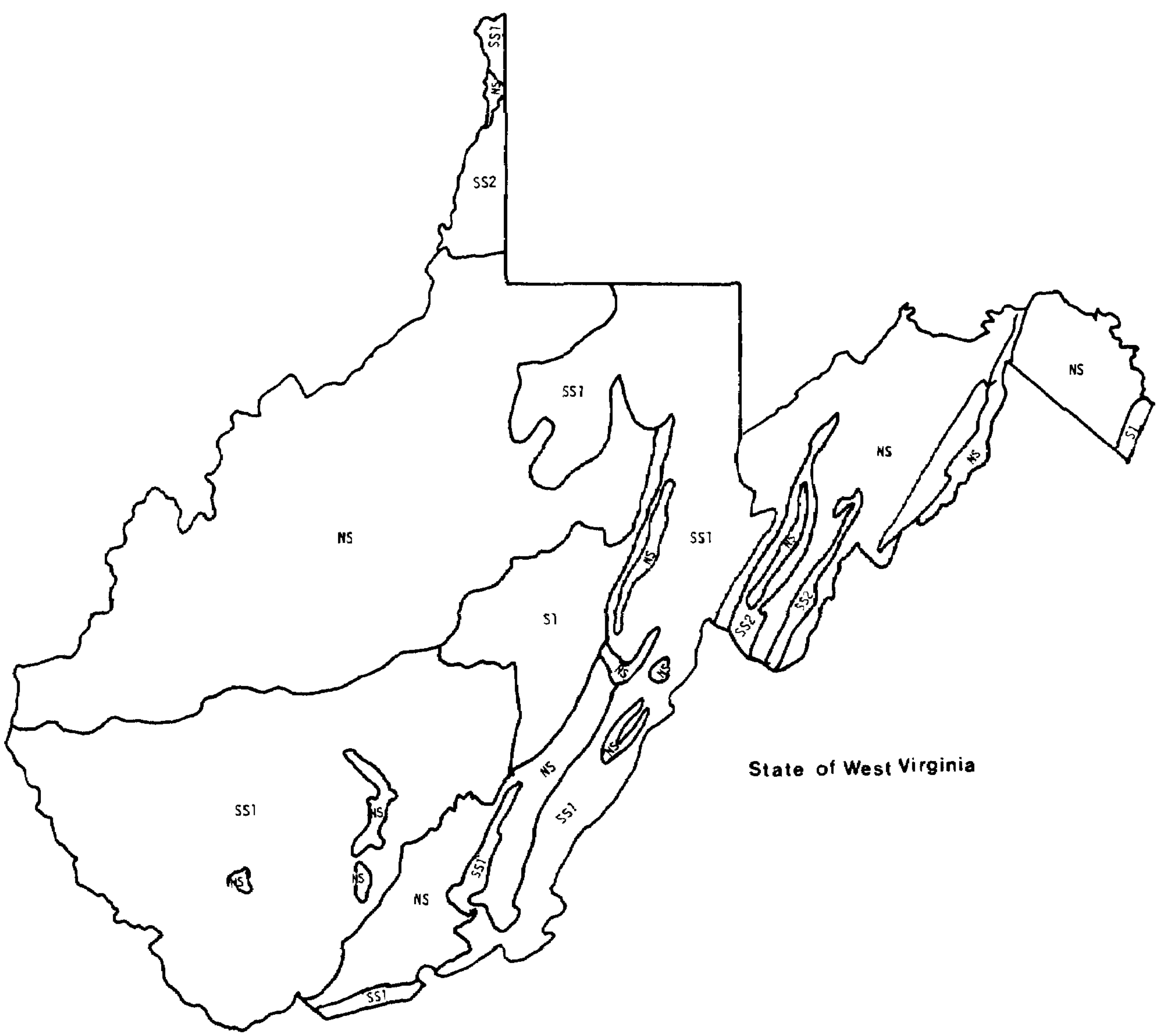


Figure 15. An example of soil sensitivity mapping based on the soil sensitivity classification of Table 12. (From McFee 1980b)

- nature of soil litter; and
- water permeability.

Sulfate adsorption is a recently identified soil sensitivity parameter of potential importance, yet more research is needed to clarify its role in the prediction of soil vulnerability. In general, it has been demonstrated that the leaching of sulfate and associated cations is dependent on sulfate mobility, or, conversely, the sulfate absorption capacity of soil (Johnson and Cole 1977; Johnson et al. 1980; Singh 1980). Sulfates in precipitation are known to pass rapidly through the top soil horizons and adsorb preferentially in the lower horizons (Singh 1980).

As a rule, excess positive charges in the soil will neutralize adsorbed negative ions such as sulfate. As soils acidify, however, deposited hydrogen ions can replace these excess soil cations, freeing them to bind with mobile sulfates. Thus, if sulfate loadings are greater than the capacity of the ecosystem to utilize or store it, sulfate leaching will occur, cations will be mobilized and the acidity of soil will increase (Reuss 1976, 1980). Moreoever, as the sulfate adsorption capacity of soil becomes saturated, further sulfate inputs will remain mobile and are subject to rapid transfer, along with associated cations, to the hydrographic network.

Sulfate adsorption in soils is enhanced by such factors as increased temperature, long equilibrium periods, decreased soil moisture, low soil pH and moderated rates of sulfate loading (Singh 1980). The presence of free iron and aluminum oxides also promotes sulfate adsorption while elevated concentrations of organic matter increase sulfate mobility by blocking available adsorption sites (Johnson 1980; Johnson et al. 1980). Sulfate mobility is difficult to predict, however, since any number of these mediating factors may co-exist in the same soil.

Large portions of the eastern United States possess soils that may be sensitive to acid precipitation (Glass et al. 1980; Klopatek et al. 1980; McFee 1980a,b). These include the shallow and steep soils of the Adirondack and Appalachian mountains as well as the coarse, noncalcareous glacial tills of New England. Moreover, considerable portions of the southeastern U.S. are located on highly weathered soils that are particularly vulnerable to the leaching effects of acid precipitation. In agricultural areas within sensitive soil regions, soil amendments and other management techniques will largely negate any adverse effects of acid deposition. In other areas of the country, the productivity of well-buffered soils may in fact benefit from excess sulfur and nitrogen deposition (National Atmospheric Deposition Program 1978).

4.4 HYDROLOGY

The hydrological characteristics of watersheds are important determinants of the sensitivity of ecosystems to atmospheric deposition. Discharge volumes, velocities and pathways relate very closely with other sensitivity factors, notably topography and soil properties. They determine rates of overland flow versus groundwater flow, the extent of soil penetration, and the residence time of waters in different components of the watershed (Norton 1980). The vulnerability of ecosystems may be enhanced by minimal soil contact, short residence times and rapid or voluminous discharges (Hendrey et al. 1980a). Polluted waters may flow essentially unaltered to groundwater and surface streams when topography is steep, soils are shallow or particularly coarse, and root channels or animal burrows are plentiful (National Atmospheric Deposition Program 1978).

Snowmelts are an important hydrological factor in ecosystem vulnerability as they may cause rapid release of the pollutants and acids that have accumulated over the course of a season. The three stages involved in the snowmelt process are all relevant to pollutant loadings in ecosystems (Johannessen et al. 1980):

- the pressing out of soil water (piston flow);
- the preferential release of accumulated ions during the first phases of snowmelt; and
- the dilution stage, where remaining water contains significantly lower ionic concentrations.

The sensitivity of aquatic ecosystems to snowmelt loading is governed by the time required for these stages to occur; rapid snowmelts are associated with more widespread and acute ecological effects. The extent of mid-winter snowmelt, with its preferential release of ions, and the degree of snowpack stratification are other important factors mediating pollutant loading of ecosystems (Wright and Dovland 1978; Overrein et al. 1980).

Furthermore, the ionic yields of snowmelt have been found to correlate inversely with amounts of snow cover (Lewis and Grant 1980b). When snow cover is sparse, soil frost is more prevalent and ionic exports to aquatic systems, especially of nitrates, are accelerated. Widespread soil frost also preferentially increases terrestrial ecosystem losses of phosphates, potassium and other nutrients of biological demand that would normally be retained by a variety of mechanisms before reaching the aquatic environment (Lewis and Grant 1980b).

4.5 HYDROCHEMISTRY

The physico-chemical properties of drainage waters largely determine their susceptibility to acidification and other alterations in water quality brought on by atmospheric deposition. As a general rule, water chemistry is a function of the geological, pedological, and vegetative characteristics of the watershed (Likens et al. 1979). The particular sensitivity of these systems more often than not parallels the degree of vulnerability observed in terrestrial components of the watershed (Likens and Bormann 1974a).

Sensitive surface waters generally possess all the characteristics of soft waters. They have low conductivities and pH values typically less than neutral. Unlike hard, mineralized waters, they are low in alkalinity and, consequently, poorly buffered.

The alkalinity of surface waters is generally agreed to be indicative of the susceptibility of aquatic ecosystems to acidification (Henriksen 1979, 1980; Hendrey et al. 1980a). Alkalinity is defined as a measure of the concentration of any available sink for hydrogen ions (Kramer 1976).

The principal buffering agent in natural waters is bicarbonate (HCO_3) or calcium carbonate ($CaCO_3$), although aluminum and other metal complexes, and organic matter, may provide some acid-neutralizing capability to softwaters (Hendrey et al. 1980a; Norton 1980). Hendrey et al. (1980a) suggest that alkalinity values less than 500 ueq/l are indicative of potential freshwater sensitivity. The Ontario Ministry of the Environment (1979) provides the following hydrochemical parameter values below which surface waters have a marked potential for acidification:

- alkalinity, 300 ueq/l (15 mg/l) of $CaCO_3$;
- conductivity, 35 umho/cm at 25° C; and
- pH, less than 6.0.

Henriksen (1979, 1980) defines water acidification in terms of alkalinity:

Acidification = pre-acidification alkalinity - present day alkalinity

Using historical water-chemistry data from 719 lakes in southern Norway, he developed the "predictor" nomograph depicted in Figure 16. It is used to predict the acid status of lakes where historical water-quality data are lacking.

This model is based on the hypothesis that acidified waters result from a large scale acid-base titration: cations released by weathering processes in the watershed are titrated with acids from wet and dry deposition (Henriksen 1979, 1980). The assumption is made that excess Ca^{2+}, or Ca^{2+} and Mg^{2+}, contained in waters reflects the weathering process while the pH of precipitation, or lake concentration of $SO_4{}^{2-}$, represent atmospheric acid inputs (all marine sources corrected for). It is also assumed that Ca^{2+} and Mg^{2+} concentrations do not change significantly in response to acidification, and that these substances are accompanied by equivalent amounts of bicarbonate (Henriksen 1980). Regression analyses led to the definition of three distinct lake classes:

- bicarbonate (HCO_3) lakes, with pH 5.3;

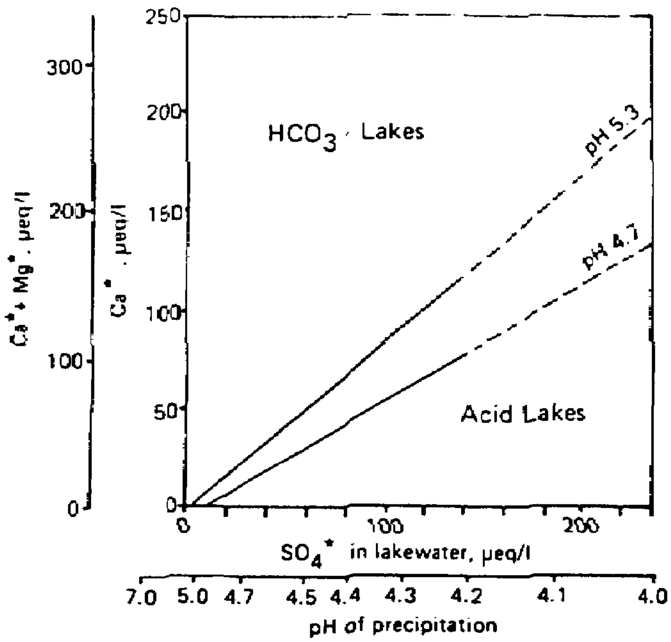


Figure 16. Henriksen's nomograph for predicting the acid status of lakes. Lake pH may be predicted from the sum of non-marine calcium and magnesium concentration or calcium content alone and non-marine sulfate concentrations in lake water or the weighted-average hydrogen ion concentration in precipitation. (From Henriksen 1980)

- transitional lakes, with pH between 4.7 and 5.3; and
- acid lakes, with pH <4.7.

The Schofield diagram, depicted in Figure 17 was developed to graphically represent the distribution of lakes within a given region according to these three lake classes.

To underscore the predictive capabilities of this model, distinct correlations have been made between judgments of fish population status and the position of lakes on the nomograph. Results from the study of 684 Norwegian lakes are presented in Figure 18. Table 14 relates the fish population status of 214 Adirondack lakes to the three lake classes, and provides an indication of the applicability of the nomograph to this region. In general, Henriksen's nomograph has been found to accurately predict the acid status of lakes in various regions of the world; an exception is found in upland lakes of Scotland where sources of acidity other than the atmosphere are thought to play a role (Wright et al. 1980). Predictive models of this nature have yet to be developed for stream acidification; at present, the measurement of water quality parameters must be relied upon for the identification of acidifying streams.

Sensitive lakes are characterized by elevated concentrations of aluminum, manganese and other heavy metals; sulfate may be the predominant anion (Wright et al. 1980). Moreover, aquatic sediments often reflect increased concentrations of heavy metals indicative of elevated dissolution processes occurring in the watershed. Metal concentrations in the water column way be augmented by decreased water pH that induces the mobilization of toxic metals bound in sediments (Kramer 1976; Gahnstrom et al. 1980). The relationship between aluminum concentrations and water pH in lakes of the Adirondack mountains is presented in Figure 19 (note log scale for Al concentrations). Reference metal levels in the water column are (Ontario Ministry of the Environment, 1979):

- < 50 ug/l for Al; and
- < 10 ug/l for Mn.

Aluminum concentrations have been found to correlate well with judgments of fish population status in acidified lakes of Norway (Overrein et al. 1980).

Freshwater acidification has been found in streams and lakes ranging from New England and New York to Florida, as well as in the Boundary Waters Canoe Area of Minnesota (Hendrey et al. 1980a). It may be anticipated in mountainous terrains of the eastern United States as well as in the Rockies and Sierras of the west (Lewis and Grant 1980a).

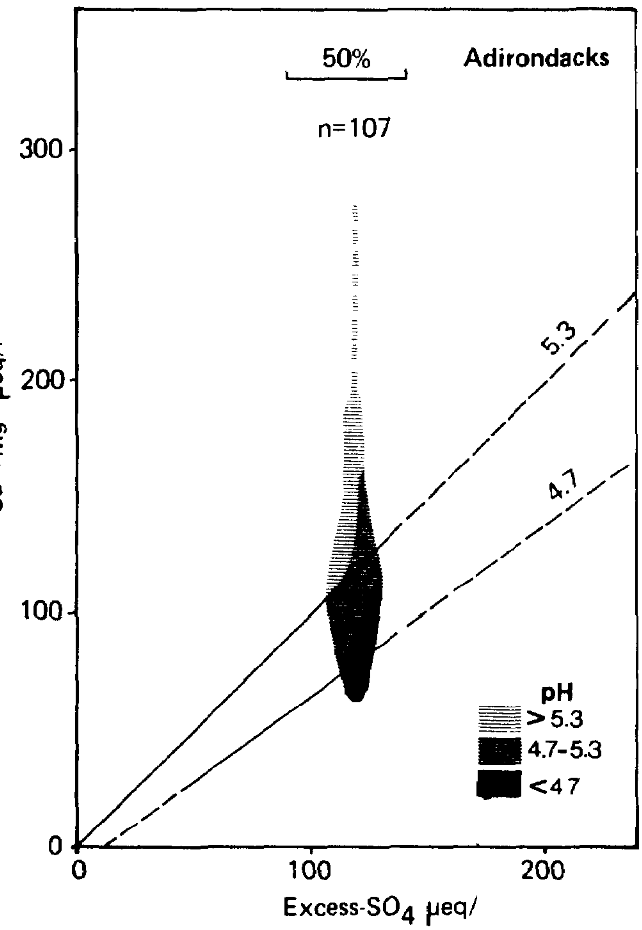


Figure 17. A Schofield diagram for Adirondack lakes, with sulfate levels of 100-120 ueq/l, superimposed on Henriksen's nomograph. The thickness of the diagram represents the number of lakes within each category of Ca^{2+} and Mg^{2+} concentration, while the shading indicates lake pH. (From Wright _et al_. 1980)

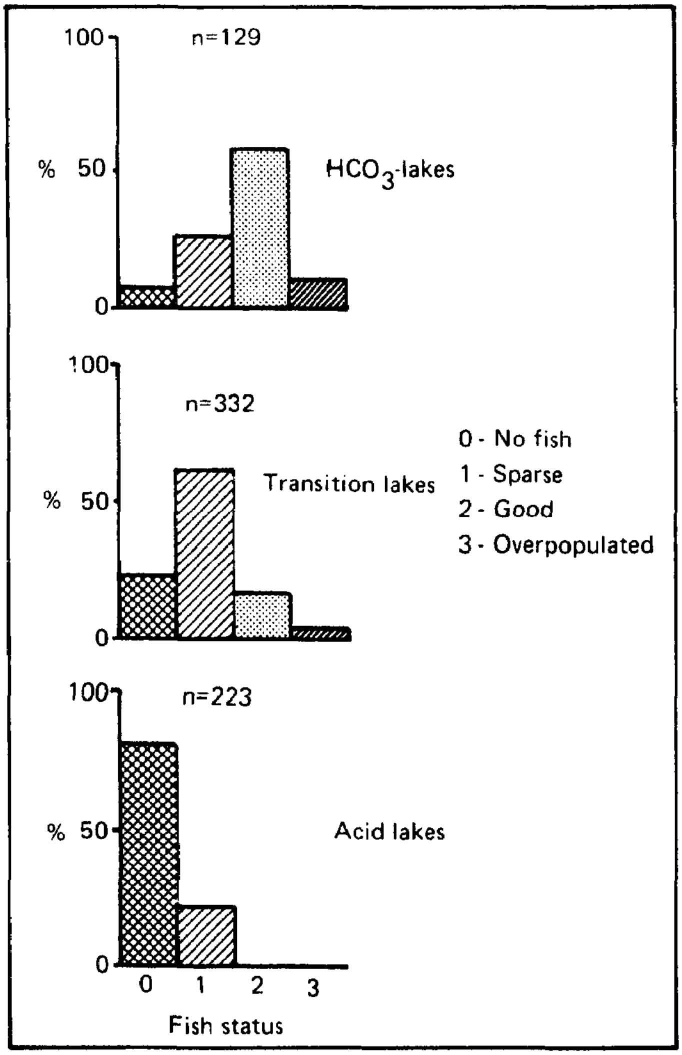


Figure 18. Frequency histograms for fish status in 684 Norwegian lakes separated according to their position on the nomograph of Figure 16. (From Henriksen 1980)

Table 14. Lake classification and fish population status of 214 Adirondack mountain lakes based on the nomograph of Figure 16.

Lake Class	Bicarbonate	Transitional	Acid
Expected pH	5.3	4.7-5.3	4.7
Calcium concentration (ueq/l)	110	73-110	73
Number of lakes	69	97	48
Number in pH range	61	65	27
Percent correctly classified	88.4	67.0	56.3
Percent of lakes without fish	2.9	54.6	91.7

(Adapted from Hendrey et al. 1980a)

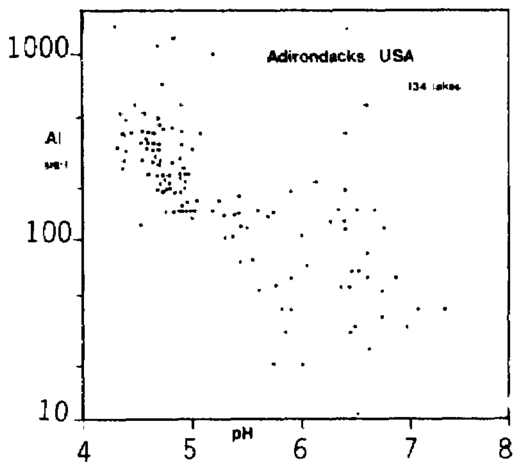


Figure 19. Total dissolved aluminum vs. pH in lakes of the Adirondack Mountains, New York. (From Wright et al. 1980)

4.6 TOPOGRAPHY

Ecosystem topography is an important, though indirect, determinant of general sensitivity to atmospheric deposition. Some relevant factors determined by topography include the size and shape of watersheds, the depth and surface area of lakes, the residence time of waters in lakes and streams, and the distribution of overland versus groundwater flow (Hendrey et al. 1980a). The ratio of watershed area to lake area and volume, for example, influences the extent of interaction between atmospheric depositions and the geological components of the watershed (Likens and Bormann 1974b). Topography also influences rates and pathways of surface run-off from precipitation events and snowmelt, especially over frozen soils (Wiklander 1980). It moderates soil depth, stability and rates of soil formation and weathering. Furthermore, it can significantly mediate regional patterns of atmospheric contaminant deposition (Wright and Dovland 1978; Schrimpff 1980).

In comparison with other ecosystems, mountainous terrains tend to induce substantially greater amounts and frequencies of precipitation than usually occur at lower elevations. As discussed above, these regions and their headwaters tend to be more sensitive to the effects of acid rain due to their smaller watersheds, thinner soils and unique meteorological conditions. For example, aquatic ecosystem acidification in Norway was first noticed in high-altitude lakes; only more recently has it been observed to occur in sensitive downstream areas (Overrein et al. 1980).

Topography also determines predominant types of vegetation, thus the nature of biotic influences on ecosystem sensitivity. A north-facing slope, for example, may support a greater number of conifers, with their characteristically acidic organic layer, while the south-facing slope may have a predominantly deciduous community, and more highly buffered soil litter.

Flatlands usually possess soils of sufficient depth and diversity to counteract the accumulation and transfer of atmospheric pollutants. Nevertheless, ecosystems situated in areas of past glaciation share many of the same susceptibilities to damage as mountainous regions, except where potential effects are modified by glacial till (Likens et al. 1979).

4.7 BIOTA

Plant cover is the major biotic factor governing terrestrial ecosystem vulnerability to air pollutants. The height, type, and density of the plant canopy directly influence the extent of gas adsorption and particle impaction. The transpiration rate of plants modifies the ratio of precipitation to evaporation in the ecosystem; when evaporation rates are elevated, acids and other foreign substances may concentrate on leaf surfaces (USEPA 1980c). Airborne pollutants may be absorbed and exchanged by plant tissues, altering the chemical composition of precipitation and other forms of deposition reaching the soil. The nature of the soil, and the biological processes which take place there, may then determine the ability of the ecosystem to retain essential plant substances.

The degree to which plant roots absorb deposited sulfates and nitrates is another biotic property determining ecosystem sensitivity. The stemflow component of incident rainfall is usually highly charged with these ions in regions of acidic precipitation, and the solution is often channeled directly to roots for uptake (Voigt 1980). These anions are most readily absorbed from soils of low nitrogen and sulfur contents, where a lack of these substances limits primary productivity. Plant uptake helps to prevent them from accumulating in soils or being rapidly transferred to aquatic ecosystems.

The composition and function of soil microflora is another biotic factor regulating the sensitivity of terrestrial ecosystems to air pollution. Certain microorganisms involved in nitrogen and sulfur conversion processes are sensitive to chemical alterations of their habitat and they may prove unable to supply sufficient amounts of the correct forms of these essential nutrients to plants (Alexander 1980a, b). Symbiotic relationships which facilitate the uptake of nutrients by plants are significant cycling mechanisms in virtually all terrestrial ecosystems, yet they are vulnerable to many different air pollution stresses (National Atmospheric Deposition Program 1978). The fungal mycorrhizae which supply nutrients and maintain the productivity of most forest trees cease to function properly at low soil pHs. A similar effect is found with the nitrogen-fixing bacteria associated with the root nodules of legumes and other free-living forms in the soil.

Microbial litter decomposition can compound the effects of atmospheric loadings by adding to soil acidity. The nitrifying bacteria, for example, routinely produce acidic nitrates through their mineralization activities (Bache 1980; Rosenqvist et al. 1980). Potential changes in species composition and dominance from exposures to air pollution or acid rain may also result in altered rates of acid or base production by soil decomposers.

The decomposition of plant residues, primarily from conifers, mosses and heather, is known to produce several organic acids which, in Norway, are believed to play an important role in the formation of acidic podzols of low base saturation (Rosenqvist et al. 1980). The litter of Scots pine, for example, has been shown to markedly augment localized soil acidification directly beneath the foliar canopy (Farrell et al. 1980).

Biota can also influence ecosystem sensitivity to atmospheric deposition through physical modifications of soil structure. Vacated root channels and the activities of soil macro-fauna, particularly earthworms and the tunneling vertebrates, all play a role in modifying the water permeability of soils (Voigt 1980). This in turn determines the degree of interaction between percolating run-off and the base exchange sites of soils.

The complexity of food webs and other expressions of species interdependence is a biotic factor determining the potential for food chain disruptions and resultant alterations in species composition. Both consumer and detritus food chains may be affected. Drastic modifications in food webs can occur when the dominant species, or those controlling energy flow in the ecosystem, are most sensitive to altered conditions in the habitat, or, as in northern climes, where food webs are simplified.

With respect to nutrient cycling in general, the degree of succession attained by the ecosystem exerts pronounced influences on the stability of individual cycles. Early successional stages, such as the flora which colonizes abandoned farmlands, are characterized by low biomass and low species diversity, and have "open" nutrient cycles. That is, system inputs and losses are elevated in comparison with storage in the biomass due to a decreased biotic control of cycling in the microhabitat. These "open" cycles are inherently unstable and perturbations, such as those brought on by air pollution and acidification, may affect ecosystem functions for a long time (Jordan et al. 1972). Late successional stages, especially on well-buffered soils, are more resistant to the effects of pollution because the essential nutrient cycles are "closed", and consequently more stable. Low inputs and losses in relation to high biomass are indicative of the ability of a productive and diverse flora to better influence its microhabitat and recover more quickly from adverse perturbations.

Ecosystems characterized by an abundance of nutrient pumps, or organisms with significant nutrient cycling capabilities, may experience increased vulnerability to the effects of air pollution if the reduced pro- ductivity of the species disrupts nutrient cycles. Rooted shoreline plants, for example, are often indispensible for the return of nutrients from aquatic to terrestrial ecosystems. Their decline has been documented in acidic waters of Scandinavia (Hendrey et al. 1976; Leivestad et al. 1976).

Ecotones, or transition communities occurring between two major ecosystems, are often sensitive to air pollution loadings. Often separating terrestrial and aquatic systems are extraordinarily productive wetlands that provide habitat for a large diversity of flora, fauna, and sensitive juvenile life forms as well as rare and endangered species. These habitats may be dominated by flora of high or low tolerance to air pollution effects, or may contain species that are subjected to additional environmental stresses such as pesticide and metal accumulation.

4.8 HUMAN ACTIVITY

Many of the human activity factors which influence the nature and extent of ecosystem loadings of airborne pollutants have been presented in discussions of pollutant emission sources, dispersion patterns, and deposition processes. Ecosystem sensitivity is in part a function of the location and extent of urban and industrial development, their relationship to prevailing winds, and the seasonality of various emissions. Figure 20, for example, compares the relation between anthropogenic sulfate loadings and decreasing pH in highly sensitive and less sensitive lakes in Sweden.

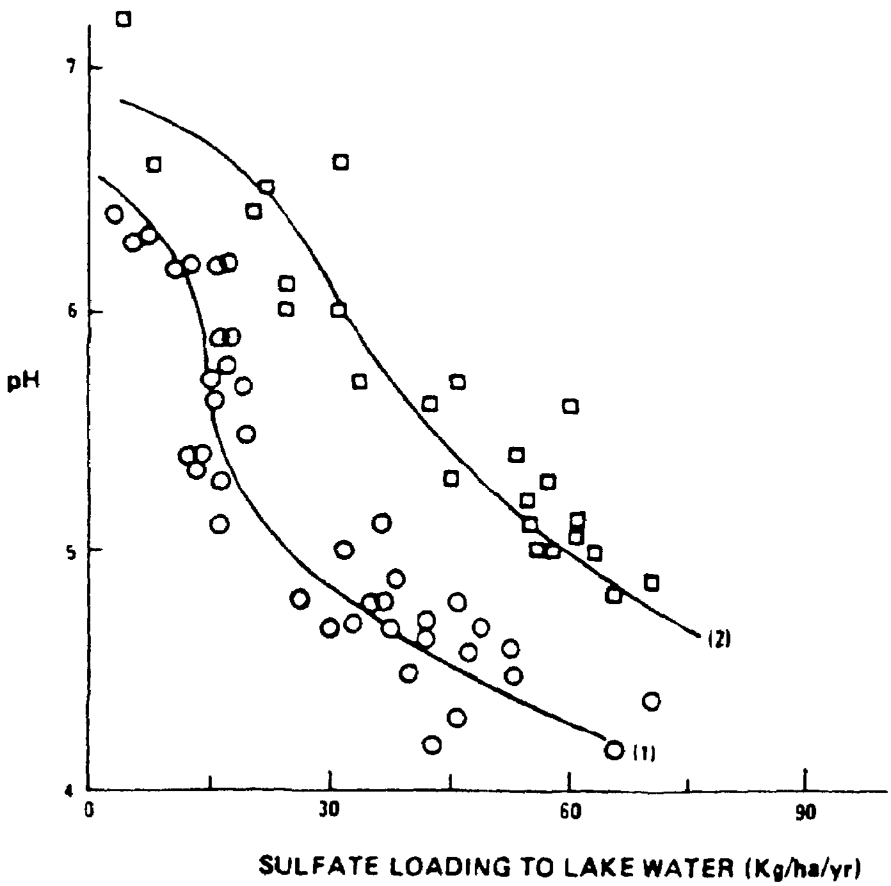


Figure 20. Data from lakes in Sweden showing the relationship between anthropogenic sulfate loadings and pH change for (1) very sensitive and (2) somewhat less sensitive surroundings. (From Glass et al. 1979)

The variety of air pollutants generated by human activities may interact synergistically to influence the degree of pollutant loadings in ecosystems. Many of these, particularly ozone, may enhance rates of sulfur dioxide conversion to sulfates in the atmosphere (GCA Corp. 1981). Moreover, the effects of acid deposition on plants, in the eastern United States and in parts of the west downwind from urban areas, are believed to differ with the intensity of exposure to ozone and other photochemical oxidants (Jacobson 1979). Point, line, and area sources may emit a variety of anthropogenic particulates that are deposited on soils; these substances may be preferentially absorbed by vegetation over natural soil constituents (Hutchinson 1979).

Land-use activities are often associated with watershed disturbances that exert considerable influence on the suspectibility of soils and surface waters to acidification and other effects of air pollution, largely through alterations in hydrology, soil structure and biota. Land use can also significantly alter the chemical composition of rain and snow (Eisenreich et al. 1980). It should be noted, however, that comprehensive studies in Norway have ruled out less disruptive former land-use patterns (dairy cattle grazing) and recent changes in land use (reindeer grazing) as contributing to lake acidification and declining fish populations (Drablos and Sevaldrud 1980; Drablos et al. 1980; Overrein et al. 1980).

4.9 SUMMARY

The sensitivity of an ecosystem to atmospheric deposition depends on its characteristic meteorology, geology, pedology, hydrology, hydrochemistry, topography, biota and human activities. The relative importance of these factors will differ from region to region. A synopsis of sensitivity factors which together render ecosystems most vulnerable to the effects of acid deposition is presented in Table 15. Areas of the United States of potential ecosystem sensitivity to acid precipitation, on the basis of soil substrata, are depicted in Figure 21.

In combination, these influences can reduce the inherent capacity of ecosystems to control short-term functions of production and biogeochemical cycling, long-term processes of succession, and the continued sustenance of fish and wildlife populations. When they occur together, they forebode serious ecological consequences for mountain ranges, pre-Cambrian terrains, first- and second-order streams, and headwater lakes. Therefore, consideration of these factors is essential to assessments of air pollution impact on fish, wildlife and their habitats.

Table 15. Factors indicative of potential ecosystem sensitivity to acidifying air pollutants.

METEOROLOGY
- Location downwind from emission sources
- Frequent precipitation, dew, frost or fog
- Precipitation exceeds evapotranspiration

GEOLOGY
- Igneous or metamorphic (alumino-silicate) bedrock
- High concentrations of aluminum, mercury and other heavy metals
- History of glaciation

PEDOLOGY
- Thin soils and organic layers (low buffering capacity)
- Mature, severely leached soils
- Low cation exchange capacity and base saturation
- Low sulfate adsorption capacity (high sulfate mobility)
- Absence of carbonates

HYDROLOGY
- Substantial snowpack accumulation
- Rapid discharges following precipitation or snowmelt

HYDROCHEMISTRY
- Oligotrophic surface waters
- Depressed water pH
- High metal concentrations
- Soft waters (low alkalinity, ionic content, conductivity and buffering capacity)

TOPOGRAPHY
- High altitude
- Steep terrain
- First and second order streams
- Headwater lakes

BIOTA
- Prevalence of conifers
- Reduced litter generation
- Early successional stage (instable nutrient cycling)

HUMAN ACTIVITY
- Watershed disturbances (deforestation, urbanization, fertilization)

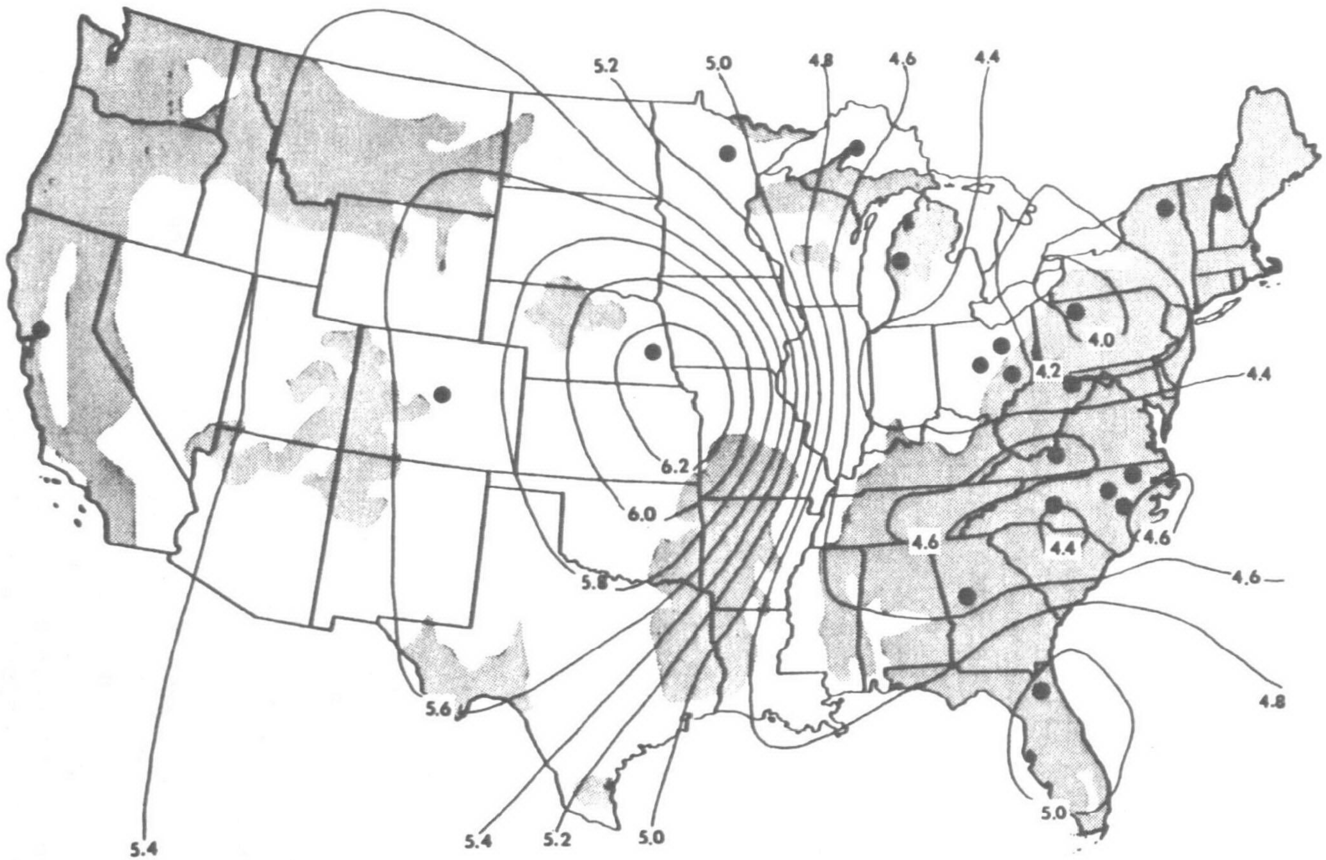


Figure 21. Areas of the United States with soils sensitive to atmospheric deposition overlain with 1978-1979 pH isopleths from the National Atmospheric Deposition Program. Black dots represent monitoring stations. (From Root et al. 1980)

5.0 RESPONSES OF FISH, WILDLIFE, AND HABITAT TO AIR POLLUTION AND ACID RAIN

The effects of air pollution and acid rain on fish, wildlife and their habitats are varied and complex. This introductory dicussion of biotic and ecosystem-level responses is designed to provide a general overview of the current state-of-knowledge regarding potential and observed air pollution impacts in terrestrial and aquatic ecosystems. For each of the three pollutant categories, photochemical oxidants, particulates, and acidifying pollutants, important research findings are highlighted and the pertinent literature is introduced. Detailed discussions of biological and ecological response, along with references to more specialized studies, are presented in the ecosystem-specific companion reports of this series.

A summary of observed air pollution effects in plant and animal life is presented in Table 16 for different levels of biological organization. Included are ecosystem-level responses that play a role in biotic impact. Although increased emphasis has recently been placed on the investigation of biotic effects in natural settings, much of the available data comes from laboratory studies performed under controlled conditions.

The responses of plants and animals to air pollution will be influenced by the structural and functional characteristics of their ecosystems. A given organism may exhibit certain physiological responses in the laboratory but responses may be different when the organism is observed in its native environment. The discrepancies between the responses of the individual organism (autecology) and those of the organism as a part of the biotic community in its environment (synecology) are due to the intervention of a number of complex interactions and modifying variables as one moves from the laboratory to the field. Perhaps the most notable of these involves the ability of the organisms in aggregate to modify the environment according to their needs (Odum 1971). Therefore, a proper study of the effects of a disturbance such as air pollution on plants, animals and their habitats must necessarily include a cautious interpretation of the results of both laboratory and field analyses.

The balance between the plant, animal, and the abiotic components of an ecosystem can be disrupted by the loss of a few species which are sensitive to air pollution leading possibly to major changes in the entire ecosystem. Indirect impacts can be quite important. For example, animals will be affected by changes in habitat and food availability resulting from air pollution injury to plants and such changes would take place even in the absence of any direct effects on the animals.

Many extensive reviews of air pollution and acid rain effects on plants and animals have been prepared. Table 17 lists a number of these with a brief description of the subjects covered.

Table 16. A hierarchical classification of biotic and ecosystem-level responses to air pollutant uptake or deposition.

Level of Organization			
Cell	Tissue or Organ	Organism	Ecosystem
Alteration of the cellular environment	Residue accumulation	Changes in growth and appearance	Pollutant accumulation in plants and animals as well as abiotic ecosystem components (e.g., soils, aquatic systems)
Changes in cellular enzymes and metabolites	Altered growth, development and energy requirements	Increased susceptibility to biotic and abiotic influences	Residue transfer and biomagnification through food chains
Alteration of cell organelles and metabolism	Altered photosynthesis, transpiration and respiration in plants	Reduced plant productivity, yield and quality	Shifts in inter- and intra-specific competitiveness
Disruption of reaction pathways	Altered blood chemistry and physiology in animals	Abnormal animal behavior	Changes in population numbers, species diversity and the spatial distribution of biota
Teratogenic, mutagenic and carcinogenic effects	Chlorosis, necrosis, death or abscission of plant organs	Altered genetic resistance	Altered birth and mortality rates in animal populations
Disruption and death of cell	Degeneration and functional disruption of lungs and other animal organs	Death of organism	Disruption of decomposition, nutrient cycling and energy transfers
			Reduction of ecosystem stability and self- regulation
			Degeneration of plant stands and animal communities

(Adapted from Guderian 1977; Newman 1975, 1980)

Table 17. Available reviews of the biotic and ecosystem-level effects of air pollution and acid rain.

Effects of air pollution on plants (illustrated)	Jacobson and Hill (1970)
Effects of air pollution on terrestrial animals	Lillie (1970), Newman (1975, 1980) and Stickel (1975)
Effects of acid deposition on terrestrial and aquatic biota (research reports and conference proceedings)	Impact Assessment Work Group (1981), Michigan State University (1981), Drablos and Tollan (1980), Hutchinson and Havas (1980), Overrein et al. (1980), Shriner et al. (1980), Toribara et al. (1980), USEPA (1980c), A.S.A.P. (1979), Evans and Hendrey (1979), Wood (1979), Hendrey (1978), National Atmospheric Deposition Program (1978), Dochinger and Seliga (1976), Giddings and Galloway (1976)
Effects of air pollution on terrestrial ecosystems	Woodwell (1970), Smith (1981)
Ecosystem structure and function (background)	Odum (1969), Likens and Bormann (1974b)

5.1 RESPONSE TO PHOTOCHEMICAL OXIDANTS

The photochemical oxidant complex constitutes a widely varying class of pollutants. The composition will depend on the nature and availability of specific precursors and the meteorological conditions prevailing during formation. The ozone (O_3) and peroxyacetylnitrate (PAN) components of the photochemical oxidant complex are known to be very toxic to vegetation in controlled laboratory and greenhouse environments (National Research Council 1977). Oxidant injury to plants is widely reported in the field as well.

Animals are affected by oxidants primarily in the eyes, lungs and upper respiratory tract (Newman 1975, 1980). Ozone dissolved in water is known to be toxic to fish. Although this phenomenon is associated with water purification systems rather than airborne oxidant contamination (Paller and Heidinger 1980), it is possible that impacts may occur to aquatic organisms frequenting the water surface in areas of chronic oxidant exposure (Taylor 1973). Since effects of airborne oxidants on aquatic organisms and environments have not been demonstrated, the following discussion emphasizes anticipated effects on terrestrial organisms and ecosystems.

5.1.1 Plant Response

Ozone is perhaps the most significant cause of air pollution damage to plants in the U.S. Plant injury has been observed in many different regions, affecting a wide range of vegetation including leafy vegetables, grains, conifers and deciduous trees.

Ozone enters the leaves of plants through the stomata during normal gas exchange. Once inside the leaf, ozone reacts with the moist cells causing injury or death of cells and tissues; preferential attack occurs on the palisade mesophyll cells. Recently matured leaves appear to be most sensitive to ozone exposure (National Research Council 1977). The response of plants can be discussed in terms of visible or subtle effects.

The visible effects of exposure to oxidants of broadleaf plants, grasses and conifers typically include (Heck and Brandt 1977):

- stipple, fleck and chlorosis on upper leaf surface, premature senescence or death in broadleafs;
- scattered necrotic areas on both leaf surfaces and necrotic streaking in grasses; and
- brown-tan necrotic needle tips and chlorotic mottling of needles in conifers.

The "emergence tipburn" disease of eastern white pine is a well-documented effect of photochemical oxidant pollution in the northeastern United States (Berry and Ripperton 1963; Skelley et al. 1979). Concentrations varying from 0.06-0.25 ppm O_3 are sufficient to produce tipburn symptoms, and primary roots are observed to die back in response (Berry and Ripperton 1963, Costonis 1970). Symptoms on this species are enhanced by excess leaf moisture common in this region (Sinclair and Costonis 1967). Other susceptible eastern species include larch, hemlock and pine varieties, while red pine, firs and spruces demonstrate greater tolerance (Davis and Wood 1973).

Chlorotic mottling of the needles is the major oxidant-related disease affecting ponderosa and Jeffrey pines in California (Taylor 1973; Williams et al. 1977; Munn et al. 1977). Ozone levels of 0.5 ppm for nine hours over 9-18 days produced these symptoms in controlled experiments (Miller et al. 1963). Subsequent research indicated that under natural conditions exposures of 0.08 ppm O_3 for 12-13 hours per day are sufficient to injure ponderosa pines (Taylor 1973). Mid-elevation pines are more severely damaged than those at higher and lower altitudes as oxidants concentrate at the ceilings of thermal inversions (Williams et al. 1977).

The quality of growing sites also influences the extent of this disease, although symptoms are found in the best growing sites. Other western species affected by oxidant exposures include white fir, sugar pine, sequoia, incense cedar, Lodgepole pine and black oak (Williams et al. 1977).

The subtle effects of ozone on plants are elicited by sublethal exposures characteristic of chronic oxidant pollution and involve interference in the normal physiological and biochemical processes (Pell 1974). The results include reduced growth and yield, closure of stomates, genetic abnormalities, reduced reproductive yield, and many more species-specific responses (Heck and Brandt 1977).

Exposure of the unicellular plant *Euglena* to ozone reduces photosynthetic rate and increases respiration (de Koning and Jegier 1968). Low-level exposures to ozone are also known to reduce the chlorophyll, protein and RNA content of duckweed species (*Lemna*), leading to destructive changes in the enzymes and membranes of cell tissues (Craker 1971, 1972). The effects of sub-lethal (0.1 ppm O_3) exposures on duckweed development include (Feder and Sullivan 1969):

- slower multiplication processes;
- lowered rates of frond doubling;
- prolonged flower production; and
- fewer flowers than controls.

Changes in photosynthesis, respiration, ATP and total adenate content in the primary leaves of pinto beans have also been correlated with the development of O_3 toxicity symptoms (Pell and Brennan 1973). Exposures of 0.3 ppm O_3 for three hours inhibited internode elongation and the initiation of new internodes after intially stimulating these parameters at lower concentrations (Leone and Brennan 1975).

Carlson (1979) showed that maple, oak and ash trees suffered reduced photosynthesis at ozone levels that do not cause visible damage. Eastern deciduous trees are generally sensitive to ozone at 0.2 and 0.3 ppm for two to four hours (National Research Council 1977). Histological and histochemical changes in the needles of ponderosa pine are also known to reduce apparent photosynthetic rates (Miller et al. 1969; Evans and Miller 1972). Much remains to be learned, however, of specific biochemical and bioenergetic alterations induced by low-level oxidant exposures on all groups of primary producers.

PAN is taken into the leaves of plants in the same manner as other gaseous pollutants, i.e., through the stomata. As opposed to ozone, PAN preferentially attacks the spongy mesophyll cells. Young, developing leaves are most sensitive to exposure to PAN (National Research Council 1976, 1977). Visible symptoms include (Heck and Brandt 1977):

- glazed, silvered, or bronzed appearance on leaf underside, bifacial collapse in a banded pattern and early senescence with abscission in broadleafs;

- irregular collapsed banding of bleached yellow to tan color in grasses; and

- needle blight with some chlorosis or bleaching in conifers.

The subtle effects on plant physiology induced by PAN exposure are similar to those discussed for ozone. PAN is especially known to preferentially inactivate enzymes with sulfhydryl groups in plants (Mudd 1963). Field studies of PAN injury are rendered impractical by difficulties in distinguishing its effects from those of ozone or total oxidant injury, therefore little plant damage has been attributed directly to PAN.

A variety of abiotic factors have been found to modify the extent of oxidant injury to plants (Heck 1968). They include:

- light intensity;

- temperature;

- relative humidity;

- soil characteristics; and

- diurnal and seasonal influences on plant physiology.

Synergism between oxidant and SO_2 exposures has been shown to occur as well; concentrations of SO_2 and O_3 that alone caused no injury brought on damage when combined (Menser and Heggestad 1966). Ozone uptake in red kidney beans was seen to increase several-fold as relative humidity rose from 35 to 75 percent (McLaughlin and Taylor 1981); oxidant injury of tobacco has been correlated with increased humidity (Otto and Daines 1969). The nutritional status of plants in part determines their susceptibility to ozone toxicity (Craker 1971). Leone et al. (1966) found that tobacco plants were most sensitive under optimum nitrogen conditions, but appeared to acquire protection when too little or too much nitrogen was present. Biotic factors regulating ozone tolerance include the age of the organism and its tissues, and inherent genetic susceptibility relative to the structure and function of stomata (Heck 1968). Biotic pathogens must be considered as well in investigations of air pollutant effects on plants. Ozone injury can enhance, inhibit or show no effect on bacterial, viral or fungal infestations, while, paradoxically, some pathogens provide localized protection to ozone injury (Heagle 1973).

Ethylene (C_2H_4) is at once a phytotoxic hydrocarbon pollutant of occasional localized importance in urban areas and a natural plant hormone that plays an important role in regulating plant development. The symptoms of ethylene injury include (Craker 1971; National Research Council 1976):

- growth reduction;
- decreased apical dominance;
- flower deformities;
- leaf growth abnormalities;
- mitochondrial swelling;
- early senescence; and
- premature leaf abscission.

Stress ethylene refers to excessive amounts of this hormone produced in response to various stresses such as air pollution (Tingey 1980). Stress ethylene production rates are found to correlate with foliar injury from ozone exposures; they accurately measure the extent of tissue response to ozone stress in plants and predict their relative sensitivity (Tingey et al. 1976). Indeed, some of the injury evidenced by oxidant exposure may be due to accelerated ethylene production by injured tissues (Craker 1971).

5.1.2 Animal Response

Information on the effects of photochemical oxidants on animals is derived mainly from laboratory studies (Lillie 1970; Newman 1975, 1980; National Research Council 1977). Ozone is the most commonly studied of the compounds making up the photochemical oxidant complex because it is known to be among the most toxic of gases. Other photochemical oxidants may be biologically active but the studies of associated effects are few. The only study of PAN reported led to the conclusion that it is less toxic to animals than ozone (National Research Council 1977). Furthermore, in laboratory studies of exposure to the complex of photochemical oxidants, the observed effects are quite similar to those of exposure to ozone (National Research Council 1977). This section therefore focuses on the known effects of ozone.

Ozone enters the vertebrate system primarily during respiration. Due to its relative insolubility in water, it can reach deep in the lungs and therefore cause damage to central airways and terminal alveoli. Ozone is a highly reactive oxidant and is likely to be absorbed by the lung tissues (National Research Council 1977). The effects of ozone depend on the length of exposure.

Generally, acute exposures to ozone have been found to cause pulmonary edema (excess fluid in the lungs), and vascular hemorrhaging in certain species. Changes in lung function have also been found to occur. These include alterations in the elastic behavior of lung tissue, increased resistance to air flow, and decreased carbon monoxide diffusion capacity (National Research Council 1977).

Several effects have been found to result from prolonged exposure to ozone. These include (Stokinger 1962):

- effects on lung tissue function and morphology;
- acceleration of lung tumor formation; and
- acceleration of aging processes.

More specific potential effects of chronic low-level exposures include (Kavet and Brain 1974):

- oxidation of structural proteins and membrane lipids;
- lung inflammation and edema;
- altered cellular enzyme activity; and
- inhibition of lung macrophage function.

Localized tolerance to ozone in the lungs is known to be acquired from previous exposures, however altered enzyme activity can irreversibly suppress the cell growth and metabolism required for adequate pulmonary function (Kavet and Brain 1974).

Ozone is also known to impair normal lung defenses resulting in increased susceptibility to infectious organisms. Enhanced susceptibility in mice has been reported at doses as low at 0.08 ppm for three hours, a level which could be achieved in rural areas downwind of major urban centers (National Research Council 1977). It is thought that ozone and other oxidants inhibit or inactivate two normal respiratory functions: the action of cilia in the nasal and upper respiratory passages that clears particles and therefore prevents them from entering the lungs, and phagocytosis by macrophages found in the alveoli (Kavet and Brain 1974; National Research Council 1977).

The differential tolerance of animal species to ozone is thought to be primarily phylogenetically controlled; nevertheless, genetic inheritance of ozone tolerance has been demonstrated as well within populations of deer mice in California (Richkind and Hacker 1980). Deer mice from areas of elevated oxidant pollution were found to be significantly more tolerant to ozone than those inhabiting areas of less pollution. Their laboratory-born progeny reflected the same trend, suggesting a genetic basis for resistance to ozone effects. Randomly bred populations are more tolerant than inbred populations, indicating the importance of genetic variability. The biochemical links have yet to be found, however, between the degree of ozone toxicity and genetic expressions of tolerance in wildlife exposed to photochemical oxidants.

A summary of photochemical oxidant effects on animals is presented in Table 18. Further investigation is required to elucidate other potential impacts of photochemical oxidants on wildlife. For example, it remains to be learned whether oxidant air pollution alters the visual and olfactory senses of vertebrate wildlife. Blindness and eye irritation are postulated effects of oxidants on bighorn sheep of the San Bernardino Mountains of California (Taylor 1973). It is also likely that oxidant alterations of specific habitats, or the ecosystem in general, may impact wildlife populations in ways that are difficult to detect or predict (Taylor 1973).

Table 18. The effects of photochemical oxidants on animals.

Ecological	Physiological
Changes in population numbers	Changes in cellular enzymes
Altered birth and death rates	Altered blood chemistry or physiology
Abnormal behavior	Lowered resistance to natural environmental stresses
Altered genetic resistance	Birth defects, mutagenesis or carcinogenesis

(Adapted from Newman 1975, 1980)

5.1.3 Ecosystem Response

Chronic photochemical oxidant pollution has been reported to produce ecosystem changes analagous to but less severe than those observed by Woodwell (1970) for radiation, heavy metal and sulfur dioxide pollution. Westman (1979) observed that, in coastal sage scrub ecosystems, chronic ozone exposures of 0.18 ppm elicited the following system-level responses:

- decreased total foliar cover;
- lower number of plant species;

- increased number of dominant species; and
- more individuals of the dominant species.

Increases in mean annual oxidant concentrations were found to correlate better than 43 other habitat variables in explaining the overall decline of foliar cover, implicating oxidant pollution as the primary stress in this ecosystem. Oxidant-induced shifts in species composition away from dominant populations have been observed in the San Bernardino Mountains of California (Miller et al. 1969), other areas of the west (Treshow and Stewart 1973; Westman 1979) and the deciduous forests of the east (Hayes and Skelley 1977; Skelley et al. 1979).

Field studies performed by Treshow and Stewart (1973) determined that oxidant levels of 0.15 ppm are sufficient to produce visible injury in the dominant species of natural plant communities. A dominant grass species was found to be the most sensitive of an oak-grassland community while the dominant aspen was most sensitive in an aspen-conifer community. In remaining plants, ozone exposure results in a general reduction of plant biomass and decreased root storage for assuring survival in following years (Price and Treshow 1972).

These effects on species composition and biomass can lead to altered nutrient cycling and energy relationships in terrestrial communities and altered hydrology and water quality in the drainage basin (Taylor 1980). Resultant patterns of succession are difficult to predict as each plant reacts in a different way to changes in community dominance. Understory species, for example, may lack the shade they require for growth in their niche and may thus not proliferate if the dominant species is selectively removed. Once changes in species composition are set in motion, the outcome is uncertain and there is little basis for a prediction of future species composition (Treshow and Stewart 1973; Harward and Treshow 1975).

Large-scale alterations in species composition and ecosystem succession may result if the oxidant-tolerant species replacing the more sensitive dominants are themselves susceptible to another environmental stress. For example, the oxidant-sensitive dominants of mountains in California are well-adapted to frequent occurrences of fire; many of the conifers that would replace them are unable to survive fires, and the net result could be a rollback of successional patterns to a shrubland stage (Kickert and Gemmill 1980).

Several specific ecosystem stresses may occur in conjunction with chronic oxidant pollution. One of the best known is the enhanced susceptibility of dominant pines in oxidant-contaminated California forests to infestation by pine bark beetles (Miller and Elderman 1977; Dahlsten and Rowney 1980). These trees are also predisposed to root pathogens (James et al. 1980). A variety of other interactions between plants and their pathogens, parasites and symbionts can occur in the presence of oxidants (Heagle 1973). Ozone, for example, is believed to inhibit the fertilization of parasitic nematodes or destroy their favored sites for plant entry, feeding or reproduction (Weber et al. 1979). Soil and root populations are protected from the direct action of oxidants yet may flourish or decline in response to altered plant metabolism or metabolite translocation.

As a general rule, oxidant damage to terrestrial habitats and changes in species composition and interrelationships may be anticipated to lead to altered population numbers and community diversity among wildlife, largely through changes in the competitive status of each species. Wildlife impacts of this nature are difficult to detect and quantify. For example, a reduction in small mammal populations of oxidant-contaminated forests in California has been reported although exact reasons are not immediately clear (Miller and Elderman 1977). The subtlety and potential extent of oxidant effects in terrestrial ecosystems remains poorly understood (Skelley 1980).

5.2 RESPONSE TO PARTICULATES

Atmospheric particulate matter is a complex mixture of natural and man-made substances that includes both primary and secondary air pollutants. The most important components of particulate matter from an ecological point of view are:

- anthropogenically generated dusts, ash, and soot;
- synthetic organic compounds;
- trace metals;
- radioactive particles; and
- non-metallic ions, including the acidic sulfates and nitrates to be discussed in Section 5.3.

The particulate composition of the atmosphere also includes mineral dusts and nutrient elements such as nitrogen, potassium, silica, sulfur, and phosphorus, which may be beneficial or essential to the productivity of terrestrial and aquatic ecosystems.

Biotic responses to particulates vary widely and depend on exposure levels and duration, affected biota, and mediating environmental conditions. As with most pollutants, acute effects are attributed to periods of high exposure while chronic effects result from long-term, low level exposures. The latter may occur as a result of long-range pollutant transport but many particulates do not have as great a range as gaseous pollutants. Trace metals and organic compounds, such as pesticides, are the particulates most often associated with chronic effects. Even when exposures remain below toxic thresholds, biomagnification in food chains can lead to toxic effects in wildlife.

5.2.1 Terrestrial Plant Response

Particulates can affect plants through external deposition on plant surfaces, or they can enter plants through opened stomata. Particulate matter deposited on the soil can also be absorbed by the root system. Heavy particulate concentrations on leaf surfaces can reduce photosynthetic activity (Heck and Brandt 1977) or block stomatal openings (Williams et al. 1971). When sufficient moisture is present, hard crusts may form on plant surfaces (Darley 1969) and leaf cuticular materials may be dissolved (USDA 1973). The exact effect on the plant will depend on the type of pollutant. Caustic particles, such as fly ash, can burn plant tissues (Dvorak et al. 1978), while the primary impact of alkaline dusts deposited on leaves will be reduced plant growth (Brandt and Rhoades 1972, 1973; Manning 1971; Lerman and Darley 1975). The result of external deposition will be disruption of normal gas diffusion and a potentially greater vulnerability to gaseous pollutants and plant pathogens (Ricks and Williams 1974). Visible effects can include necrosis, chlorosis, and stunting or deformation (Dvorak et al. 1978).

Some particulate matter, metals for example, are readily absorbed by leaf and root systems. Depending on the plant and the metal, absorption may take place preferentially through the aerial or underground portions of the plant and metal translocation within the plant may or may not occur. For example cadmium is absorbed with greater efficiency by plant roots than by leaves (Haghiri 1973). It is rapidly translocated to the aerial portions of plants, although reproductive organs have been reported to accumulate less Cd than vegetative tissues (Pietz et al. 1978). Although foliar absorption of lead is the chief mechanism of accumulation in the aerial portions of plants, lead absorbed through plant roots may be translocated to above-ground plant parts (Motto et al. 1970; Lagerwerff et al. 1973).

Many particulates are accumulated and magnified in food chains. Lichens and mosses are well-known accumulators of atmospheric metals and have been intensively investigated for their potential in monitoring rates of trace metal deposition. Table 19 lists some important references in this area. Mondano and Smith (1974) showed that epiphytic mosses as well as tree needles and twigs accumulate mercury to levels exceeding those found in soils. The leaves and twigs of urban woody trees have also been observed to accumulate excessive amounts of atmospheric metals (Smith 1972, 1973). Within two kilometers of a zinc smelter, tree parts have been found with zinc levels of 4500 ppm and Cd levels of 70 ppm (Buchauer 1973). Metal accumulations usually render plants unsafe for animal consumption.

Table 19. Selected references on the use of lichens and mosses in monitoring the deposition of atmospheric trace metals.

Goodman and Roberts 1971	Gough and Erdman 1977
Grodzinska 1978	Little and Martin 1974
Groet 1976	Pilegaard 1979
Mondano and Smith 1974	Rasmussen 1977
Pilegaard et al. 1979	

The degree of particulate accumulation will depend on a number of varying factors. Plant leaf morphology is perhaps the most influential biotic factor mediating particulate accumulation on plant surfaces (Little and Martin 1972). Once on the surface, penetration will depend on the environmental conditions which govern the opening of stomata. Soil chemistry will also influence root uptake. For example:

- cadmium uptake through plant roots is enhanced in acid soils with low levels of organic matter to adsorb metals (Andersson and Nilsson 1974);
- similar conditions in the presence of soluble lead facilitate uptake of lead (Zimdahl 1976);
- the uptake of certain metals may be reduced by the presence of other metals in the soil (McGrath et al. 1980);
- increased soil acidity can facilitate plant uptake of fluorides accumulated in soils (Thompson et al. 1979).

Metal toxicity in plants usually results fron interference with respiration and photosynthesis caused by the binding of metals to chloroplasts and mitochondria (Zimdahl 1976). Cadmium, for example, accumulates preferentially in chloroplasts, reducing growth and causing chlorosis, leaf wrinkling, and red veins (Barber and Brennan 1974). Cadmium is acutely toxic in small quantities; 2.5 ppm in soils is sufficient to produce toxicity symptoms (Haghiri 1973).

Toxicity varies for different metals. Lead toxicity is generally reduced in plants by the immobilization of the metal in structural tissues (Rains 1971; Zimdahl 1976). While such mechanisms may reduce plant injury, secondary effects can take place when lead-contaminated plants are eaten by animals. Fluoride particulates are well-known phytotoxins whose impacts have been observed primarily in forest ecosystems of the northwestern United States (Miller and McBride 1975; Weinstein 1977). The toxicity of fluorides is related to their solubility in water; thus gaseous fluoride is generally more toxic than particulate forms (Jacobson and Hill 1970). Moreover, the symptoms of fluoride injury may differ from one ecosystem to another for the same plant species (Amundson and Weinstein 1980).

The threshold and extent of plant damage can be modified by a number of factors. Nutritional deficiencies, meteorological conditions, and soil variables affect the concentrations of metal required to produce toxicity symptoms (Zimdahl and Arvik 1973; Zimdahl 1976; Gough et al. 1979). Stress ethylene production is stimulated by soil applications and may influence the extent of plant damage of Cd, Pb, and other metals (Rodecap and Tingey 1981). Metal-induced foliar injury is enhanced in the presence of SO_2 although the interaction of SO_2 and trace metals does not appear to affect metal uptake rates (Krause and Kaiser 1977).

5.2.2 Terrestrial Animal Response

Animals may be exposed to particulate pollution directly through inhalation or indirectly by ingestion. While some particulate matter is biologically inert, other forms of particulates are directly toxic or may adsorb or contain toxic components. Animal responses will vary widely depending on the pollutant, the exposure pathway, and the animal's tolerance, life habits, and position in the food chain. The response of animals to particulates is discussed by Newman (1975, 1980) and Gough et al. (1979). An indication of the diversity of the effects is given in Table 20, which summarizes some retrospective observations of wildlife damage in the field.

Atmospheric particulates, notably the trace metals, fluorides, and pesticides, bioaccumulate in both vertebrates and invertebrates. The field studies listed in Table 21 document bioaccumulation of metals in wild populations. These studies found different rates of trace metal accumulation among individuals, species, and groups of related species. The most prevalent target organs for trace metal accumulation are the bones, kidneys, liver and brain. Known target organs for constituents of atmospheric particulates are given in Table 22.

The accumulation of non-essential metals, such as lead and cadmium, increases with age in vertebrates. Biologically essential elements, on the other hand, are maintained at stable levels through physiological regulation (Schlesinger and Potter 1974). In addition to age, the life habits and nutritional requirements of each species will be important determinants of particulate uptake and bioaccumulation. This has been demonstrated for small mammals in a number of studies (Quarles et al. 1974; Getz et al. 1977b, ; Clark 1979; Wright et al. 1978). Inter-species variability in accumulation has been clearly shown. Grasshoppers do not accumulate roadside lead while levels of lead in earthworms near roads correlate with the lead content of the soils (Van Hook 1974; Scanlon 1979). Earthworms are known to have a high affinity for cadmium (Ireland 1979). In general, this metal is accumulated much more rapidly than lead or zinc in soil invertebrates (Scanlon 1979).

Table 20. Incidents involving the adverse effects of atmospheric particulates on vertebrate wildlife.

Location	Species	Pollutant(s) and source(s)	Reported atmospheric levels or tissue concentrations	Effects	References
Montana, USA	Mule and white-tail deer	Fluoride, aluminum plant	Up to 430 ppm in vegetation	Fluorosis	Kay _et al_. (1975)
Washington, USA	Black-tailed deer	Fluoride, aluminum	3,900 ppm in metatarsals	Fluorosis	Newman & Yu (1976)
Ontario, Canada	White-tailed deer	Fluoride plant, industrial complex	Up to 7,125 ppm in bone and 1,200 ppm in water	Fluorosis	Karstad (1967)
England	Sparrowhawks and song thrushes	Cadmium, smelter	Up to 387 ppm in kidney	Food chain magnification	Martin & Coughtrey (1976)
California, USA	Voles	Lead, urban sources	1.1 ppm in bone	Biological contrations	Hirao & Patterson (1974)
New Hampshire, USA	Mice and shrews	Lead, cadmium, and copper, industrial regions	Body conc. for Pb, Cu, and Cd of 2.7, 3.2, and 0.4 ug/g in mice, 2.6, 2.9, and 0.4 ug/g in shrews	Biological concontrations	Schlesinger & Potter (1974)
England	Wood mice and bank voles	Mercury, chloro-alkali plant	37 to 124 $ng/dm^2/d$ deposition	Biological concentration	Bull _et al_. (1977)
Czechoslovakia	Hares	SO_2 and fly ash, power plants and other industries	0.15 mg SO_2/m^3 in air and 300 $t/km^2/yr$ fly ash deposition	Hypocalcemia and hypoproteinesis	Novakova & Roubal (1971) Novakova _et al_. (1973)

(Adapted from Newman 1975, 1980)

Table 21. Selected references on the bioaccumulation of trace metals in wild animal populations.

Pb	Clark 1979 Getz *et al.* 1977a, 1977b Goldsmith and Scanlon 1977 Hirao and Patterson 1974 Jeffries and French 1972 Mierau and Favara 1975 Ohi *et al.* 1974 Tansy and Roth 1970 Ward and Brooks 1978 Welch and Dick 1975 Williamson and Evans 1972
Fluoride	Wright *et al.* 1978 Carlson and Dewey 1971 Dewey 1973
Synthetic organic compounds	Stickel 1975
Hg	Bigler and Hoff 1977 Bull *et al.* 1977
Cu and Ni	Ranta *et al.* 1978
Pb, Cd and Cu	Schlesinger and Potter 1974
Pb, Cd and Zn	Ireland 1979 Johnson *et al.* 1978 Martin and Coughtrey 1976 Scanlon 1979 Van Hook 1974

Table 22. The major biological systems of animals affected by atmospheric particulates.

Atmospheric particulates	Major biologial systems of animals: Central nervous system	Circulatory system	Gastrointestinal system	Hepatic system	Pulmonary system	Respiratory system	Renal system	Skeletal & dental system
Asbestos					●			
Arsenic			●			●		
Barium	●				●			
Beryllium						●		●
Boron	●			●		●		
Cadmium		●		●	●		●	
Chromium			●			●		
Fluoride							●	●
Iron						●		
Lead	●		●					
Manganese	●				●			
Mercury			●	●		●	●	
Molybdenum			●					
Nickel					●			
Selenium	●		●					
Vanadium		●			●	●	●	
Zinc			●					

(Adapted from Newman 1975, 1980)

The tolerance of animals to the accumulation of particulate matter varies widely. Many species of terrestrial animals accumulate lead and other particulates to high levels with no apparent ill effects. Quarles et al. (1974) observed lead levels in small mammals that would produce severe symptoms of lead poisoning in humans and other higher vertebrates at similar body-tissue concentrations.

Particulate exposure and accumulation can cause effects in animals ranging from irritation to debilitating disease, reproductive failure and death (Lillie 1970; Newman 1975). When inhaled, many types of particulates, in particular the fine particulates, are deposited on the respiratory surface of the lungs (alveoli) where they can cause symptoms ranging in severity from slight difficulty in breathing, to emphesyma and lung cancer (Lillie 1970). In sufficient concentrations, they are known to depress the viability of protective lung-macrophage cells (Zarkower and Ferguson 1978).

In addition to the effects on lungs, many types of particulate matter are directly toxic. For example, the chronic effects of synthetic organic compounds on the reproductive success of wild bird species are well documented (Stickel 1975). Several metallic compounds present as pollutants have been observed to cause toxic, carcinogenic, and/or reproductive effects in experimental animals (Lillie 1970; Newman 1975).

Bacterial screening tests for mutagenicity have been applied to several types of particulates and materials which adsorb to particulates. Substances which demonstrated mutagenic potential include:

- fly ash (Fisher et al. 1979);
- filtrates of fly ash (Chrisp et al. 1978);
- diesel soot (Li and Royer 1979);
- polycyclic aromatic hydrocarbons (Glass 1979b);
- uncharacterized substances in urban aerosols (Alfheim et al. 1980); and
- organic micropollutants undergoing long-range transport (Alfheim and Moller 1979).

The effects of atmospheric fluoride particles are well-documented. Fluorosis, a disease causing deterioration of the bones and teeth, occurs in livestock and wild vertebrates as a result of exposure to fluorides and subsequent bioconcentration (Karstad 1967; Kay et al. 1975; Newman and Yu 1976; Krook and Maylin 1979). The concentration of fluoride in the femur is believed to be the best indicator of uptake in vertebrates (Wright et al. 1978). Reductions of arthropod populations have also been attributed to fluoride bioaccumulation (Carlson and Dewey 1971; Dewey 1973).

Many sub-lethal effects can result from exposures to atmospheric particulates. The ingestion of alkaline dusts, for example, can change stomach and intestinal pH in wildlife and livestock, potentially altering the composition of internal microflora, or caustic ash may irritate stomach linings and predispose the animal to infection or produce symptoms of gastro-enteritis (Tendron 1964). Atmospheric lead can depress levels of specific enzymes in the blood of pigeons (Ohi et al. 1974). Combined particulate and sulfur dioxide pollution has been shown to effect blood proteins, serum chemistry and urine pH, as well as reproductive success in hares (Novakova 1970; Novakova and Roubal 1971; Novakova et al. 1973). Table 23 summarizes the variety of responses anticipated from the exposure of wildlife populations to particulate air pollutants.

5.2.3 Terrestrial Ecosystem Response

The effects of intrinsically toxic particulates on biota can lead to disruptions of ecosystem functioning both in the vicinity of point sources, where exposures may be acute, and in areas of chronic, low level deposition. Observed effects include changes in soil microbial activity, nutrient cycling, species growth and diversity, succession, and the integrity of food chains. Furthermore, the effects of widespread ecosystem contamination from atmospheric particulates are probably not easily reversed (Purves 1972).

Jackson and Watson (1977) studied the effects of lead, cadmium, copper, and zinc emitted from a lead smelter and identified two distinct stages of ecosystem disruption corresponding to the amount of pollutant loading. In a far zone, 1.2-2.0 miles from the stack, soil and litter microfauna were unaffected but forest litter was observed to accumulate. In this zone, the nutrient pool size was elevated by litter accumulation.

Closer to the smelter, in a zone 0.4-0.8 miles from the stack, reduced decomposer communities were observed. Here deposited metals accumulated at a rate too great to be assimilated. High concentrations of metals in leaf litter inhibited decomposition by soil microflora and the size and quality of the nutrient pool was depressed (Jackson and Watson 1977).

Under the more severe conditions observed in the near zone, nutrients tend to remain bound in dead tissue. Soil acidity increases and the cation exchange capacity decreases as a result of reduced decomposition, leading to the mobilization and translocation of free soil nutrients, notably calcium (Jackson and Watson 1977; Freedman and Hutchinson 1980a, c, d). Reduced availability of nutrients for plant uptake can result in a decreased nutrient content of living biomass and future forest litter. In sum, mineralization and decomposition rates are depressed at the same time that nutrient losses from the ecosystem are accelerated.

Table 23. Documented responses of wildlife, domestic and laboratory animals to acute and chronic exposures of atmospheric particulates.

Responses	Selected air pollutants: Arsenic	Asbestos	Barium	Beryllium	Boron	Cadmium	Chromium	Fluoride	Iron	Lead	Manganese	Mercury	Molybdenum	Nickel	Selenium	Vanadium	Zinc
Changes in population numbers	●	●				●		●									
Changes in blood chemistry or physiology	●			●		●				●			●	●		●	
Changes in cellular enzymes	●			●				●	●								●
Changes in external appearance	●		●					●		●			●		●		
Change in population distribution								●									
Change in death rate (in *free living animals*)	●	●		●	●	●		●		●					●		
Change in birth rate	●									●							
Change in growth rate				●				●		●							
Change in genetic resistance															●		
Abnormal behavior	●		●	●	●		●	●		●	●	●	●		●	●	●
Physiological changes observed in autopsy and histological analysis	●	●				●		●		●	●			●	●	●	●
Lowered resistance to natural environmental stress	●							●									
Residue accumulation in body tissues	●	●		●	●	●	●	●	●	●	●	●	●		●	●	●
Teratogenic, mutagenic, or carcinogenic effects		●	●	●		●	●							●	●		

(Adapted from Newman 1975, 1980)

As discussed above, depressed rates of decomposition are important as an indicator of soil response to atmospheric pollutant loading. Reductions in microbial activity have been documented in a number of ways:

- decreased weight loss in forest litter samples (Inman and Parker 1978);

- depressed CO_2 evolution, soil respiration, substrate decomposition, and enzyme activity (Ruhling and Tyler 1973; Ebregt and Boldwijn 1977; Freedman and Hutchinson 1980a, d);

- carbon nitrogen ratios in litter are inversely proportional to microbial populations and their activity (Jackson and Watson 1977); and

- amino-sugar carbon concentrations correlate negatively with the presence and function of both fungi and soil arthropods (Jackson and Watson 1977).

The ecosystem responses to elevated metal deposition are reductions in species diversity and altered plant succession. Gilbert (1975) reported drastic changes in forest structure due to fluoride contamination while Freedman and Hutchinson (1980b) documented reduced tree diversity and biomass in forests near smelters. Desert grassland communities have been observed to increase in species diversity with increasing distance from point sources of copper and other metals (Wood and Nash 1976; Dawson and Nash 1980). Dust accumulations in eastern deciduous forests have also provoked shifts in species dominance and community composition (Brandt and Rhoades 1972, 1973).

The observed shifts in plant dominance and diversity may in part be due to pollution-induced changes in pathogen and symbiont communities. Some particulates are known to inhibit plant pathogen activity, for example, pine rust (Parmeter and Uhrenholdt 1975), while other infectious organisms, notably fungi, may be stimulated (Manning 1971). Fluorides have been observed to predispose natural plant communities to insect attack (Carlson _et al._ 1974).

Biomagnification of substances contained in atmospheric particulates is an important ecosystem response. Well-known examples of biomagnification in terrestrial food chains include radioactive particles in reindeer and pesticides in piscivorous and carnivorous birds (Stickel 1975). Food chain biomagnification probably contributes to the elevated concentrations of trace metals observed in small mammals and birds of intermediate trophic levels (see references in Table 21).

The tendency for substances to biomagnify may be reduced by natural purification mechanisms which can efficiently remove some accumulated particulates from food chains. For example, in terrestrial ecosystems there are at least four obstacles to Pb biomagnification in food chains (Jones and Clement 1972):

- the immobility of Pb in soils;
- restricted translocation from roots to shoots;
- restricted absorption of ingested lead by herbivores; and
- the preferential retention of Pb in inedible bone tissue.

Hirao and Patterson (1974) studied lead aerosol pollution in remote areas of the California Sierras. They confirmed that while herbivores tended to bioaccumulate Pb from leaf surfaces, it occurred in such a way that Pb was not biomagnified at higher trophic levels. Natural purification mechanisms involving snowmelt interactions with contaminated forest humus prevented further Pb uptake by prey organisms at the base of food chains.

Such processes may not be effective barriers for other types of particulate matter. Moreover, different ecosystems will have varying mechanisms and capacities to protect food chain integrity. Should these be non-existent or easily circumvented by certain metals and organics, food chain biomagnification can readily occur.

The ecological effects of particulates appear to be most severe when acidifying pollutants, especially sulfur dioxide, are present, although few efforts have been made in field studies to distinguish the relative influence of each. Biotic effects of this synergism are not well known, however atmospheric metal deposition in combination with acid precipitation can cause fundamental changes in soil structure and its organic content: cation exchange capacity and pH are both reduced, aluminum and other cations are mobilized, and soil erosion is promoted in areas of significant forest destruction (Hutchinson and Whitby 1974). All of these conditions can substantially increase pollutant phytotoxicity, and impact water quality and hydrology, exacerbating the effects of particulates on aquatic biota.

5.2.4 Aquatic Plant Response

Impacts on aquatic ecosystems related to particulate pollution are, for the most part, associated with trace elements, especially metals. Water bodies accumulate metals from a variety of sources. Particulate matter may be deposited directly in aquatic ecosystems through both wet and dry removal processes, or matter deposited in the watershed may run off into surface waters (Wright and Gjessing 1976; Galloway and Likens 1979; Overrein et al. 1980; Schindler et al. 1980a; Eisenreich et al. 1981). Trace elements may also be derived from natural weathering processes in the catchment or mobilized from sediments (Likens and Bormann 1974b; Norton 1980; Van Hassel et al. 1980).

Releases of terrestrial and sediment-bound metals can be greatly accelerated by acidic conditions in the watershed (Malmer 1976; Driscoll 1980; Gorham and McFee 1980; Norton 1980; Schindler et al. 1980a). In fact, it is hardly possible to discuss the effects of atmospheric particulates, and trace metals in particular, without considering the effects of acid deposition (Section 5.3 of this report). Usually both types of pollution are present together in field exposure situations. In addition the acidification of soils and water has a strong effect on the mobility of trace metals and hence the biological availability of these potential toxins (Wright and Gjessing 1976; Almer et al. 1978; Henriksen and Wright 1978; Driscoll et al. 1980; Schindler et al. 1980a).

Studies of trace metal effects on aquatic flora must necessarily incorporate the synergistic effects of habitat acidification (Van Loon and Beamish 1977). For example, Stokes et al. (1973) suggested that trace metals had adversely affected the phytoplankton populations of two lakes located near smelters. It was later concluded, however, that the observed effects were probably due to the combination of excessive metal concentrations with low water pH (Stokes and Hutchinson 1976).

Yan (1979) compared the phytoplankton communities of uncontaminated lakes to those of partially acidified, metal-contaminated lakes. The changes in community composition which were found strongly resembled those observed in acidified waters with low metal concentrations (Section 5.3.4). These included the replacement of dominant chrysophyte genera with a single dinoflagellate species, and a significant reduction in the number of taxa with no concurrent decrease in biomass (Yan 1979). The overall alteration may be summarized as one of:

- species replacements and shifts in dominance;
- decreased number of species; and
- expanded populations of remaining species.

Aquatic macrophytes are also subject to impact from atmospheric metals, the synergistic influence of acidification and antagonistic or synergistic effects of combinations of trace elements (Dvorak et al. 1978). Gorham and Gordon (1963) first reported that trace metal levels of toxicity to aquatic vegetation are achieved in lakes near the Sudbury, Ontario, smelters as a result of emissions. Both submerged and emergent forms are known to effectively bioaccumulate trace metals from surrounding water and sediments (Tiffen 1977). Cladophora species of algae and the cat-tail Typha accumulate the highest levels, although internal concentrations are found to vary among the different organs of the same plant (Wells et al. 1980). Algae may also play a significant role in the mobilization of sediment-bound metals as they possess the ability to sorb and accumulate them in living tissues (Laube et al. 1979).

Trace element contamination in the absence of freshwater acidification has been documented near roads. Van Hassel et al. (1980) were able to correlate stream sediment concentrations of lead, nickel and zinc to the traffic density of adjacent roads. Sediments were postulated to be the primary source and storage reserve of the bioaccumulating trace metals in these streams, yet no toxic effects due to bioaccumulation were observed in aquatic flora.

The duckweed, Lemna sp., demonstrates acute sensitivity to elevated metal concentrations and has been suggested as a viable bioindicator for detecting metals in aquatic habitats (Nasu and Kugimoto 1981). Growth inhibition, measured as a reduction in frond multiplication, can be detected at cadmium levels as low as 0.01 ppm in water, while death occurs at cadmium concentrations of 0.5 to 1.0 ppm (Hutchinson and Czyrska 1975). These plants bioaccumulate lead, cadmium and copper to levels nearly a thousand times greater than those in water. Many other trace elements are accumulated to a lesser degree by this species.

5.2.5 Aquatic Animal Response

Aquatic fauna, from invertebrates to fish, are among the wildlife populations most affected by environmental exposures of atmospheric particulates, especially in areas also receiving acid precipitation. Both the organic micropollutants and trace elements bioaccumulate to varying degrees in freshwater organisms. Detailed reviews of the effects of trace elements in aquatic organisms have been prepared by Douderoff and Katz (1953), Katz (1975) and Sauter et al. (1976).

The extent to which aquatic fauna are contaminated by organic micropollutants from atmospheric deposition is not well determined in North America. In Norway, where greater concentrations of PCB and DDE have been found in the flesh of fish from southern lakes than in those of more northern regions, studies of long-range transport and deposition patterns of these particulates in Norway suggest atmospheric deposition as the major source (Lunde et al. 1977; Overrein et al. 1980). Little is known of the precise physiological effect in fish bioaccumulating these substances, however, they are known to undergo biomagnification in food chains and many have the potential to adversely affect reproduction in higher-order piscivores (Stickel 1975).

Many of the trace elements are potentially toxic to aquatic fauna, causing severe injury and death when present at elevated concentrations. Sublethal exposures can impair fish and other organisms by reducing their growth and reproductive success. However, as noted above for the case of plants, it is often difficult to separate the influence of most individual trace elements from that of acid deposition in aquatic communities (Gorham 1976). It is also difficult to identify the effects of single elements since effluents usually contain several types of contaminants, leading to the possibility of additive, synergistic, or antagonistic effects. A notable exception is the ubiquitous trace element aluminum which is not a significant component of atmospheric deposition, but rather is leached from the watershed in sensitive areas receiving acid deposition (Abrahamsen et al. 1979; Johnson, N. 1979; Driscoll et al. 1980). This terrestrial leachate will be discussed in connection with acid precipitation in Section 5.3.

Mercury is a toxic trace metal found as an atmospheric particulate. Concentrations of mercury can be elevated in areas receiving acid precipitation. Local atmospheric emissions have been correlated with the pH of lakes in Sweden as well as with elevated mercury concentrations in pike (Jernelov 1980). Rain and snow in Quebec, Canada, are known to scavenge 20-100 mg/l total mercury and deposit it in surface waters with background concentrations of only 5-10 mg/l (Tomlinson et al. 1980). These studies have shown atmospheric deposition in both Canada and Scandinavia to be the major source of mercury in unbuffered waters not already subject to point discharges of this metal. Most of the research on mercury in aquatic ecosystems is focused on exposure pathways and bioconcentration, with relatively little defining the effects on aquatic organisms.

Freshwater acidification is an important controlling mechanism of mercury toxicity as it determines the availability of the toxic monomethyl form (CH_3Hg). Elevated concentrations of this form are found in freshwaters, primarily in spring following snowmelt, however other aspects of acidification may account for increasing levels (Jernelov 1980; Wood 1980):

- waters of low pH and conductivity retain more mercury from precipitation than well-buffered waters;
- acidified waters revolatize less mercury than waters of higher pH; and
- greater amounts of mercury are transformed to the monomethyl form at pH levels below 7.0, either through increased bacterial production or through interactions between inorganic mercury and the humic and fulvic acids of freshwaters.

As a result, higher rates of mercury uptake are found in fish of low pH waters than in fish of well-buffered waters (Scheider et al. 1979). Several factors have been postulated to enhance mercury uptake in fish under conditions of acidification (Jernelov 1980; Tomlinson et al. 1980; Wood 1980):

- monomethyl mercury, like the organic micropollutants is lipophilic; due to this property and its high affinity for sulfur-groups present in blood, it is rapidly transferred through gill membranes;
- decreased food availability in an acidified environment results in more water being drawn through the gills, hence a greater mercury uptake per unit increase in weight;

- decreased biomass per unit of water volume, also an effect of acidification, results in greater mercury availability per unit weight of remaining biomass, and individual uptake is more rapid;

- acidification may result in the eradication of entire year classes of fish, meaning that predators are likely to receive greater amounts by consuming older, more contaminated prey.

Mercury tends to accumulate in fish so that body levels are greatest in the older individuals of a population; levels also correlate with fish length. Mercury is also subject to biomagnification in fish at higher trophic levels. For example, smallmouth bass in the Adirondacks were found to accumulate mercury at greater rates than white suckers because of their higher metabolic rate and differing food preferences (Bloomfield et al. 1980). The tendency of mercury to biomagnify in food chains was confirmed in a study by the Michigan Water Resources Commission (1972) which reported that predatory fish of the Great Lakes have mercury concentrations double those of bottom feeders. Of all trace metals assayed in this study, only mercury was found to bioconcentrate in fish to levels potentially harmful for secondary and tertiary consumers. Pike (Esox sp.) are reported to be sensitive indicators of environmental contamination by mercury as they concentrate the element in body tissues to levels 3000 times higher than surrounding water (Johnels et al. 1967).

Other trace metals affect aquatic fauna but wide differences have been observed between metal levels causing fish mortality and those producing sublethal stress effects or reduced growth (Sauter et al. 1976). Continuous low-level exposures of copper (Cu) found to physiologically impair fathead minnows were only 3-7 percent of the 96-hour median tolerance limit for adults (Mount 1968). Reproduction in the fathead minnow was shown to be inhibited at zinc (Zn) levels that had no effect on the survival, growth, and maturation of eggs and fry (Brungs 1969).

Few adverse effects on fish have been observed at levels below 10 ug/l Cu and 90 ug/l Zn in metal-contaminated lakes, although bottom-feeders can be exposed to greater amounts and be affected at these water levels (Van Loon and Beamish 1977). White suckers in lakes contaminated with zinc and cadmium (Cd) were observed to grow faster at maturity and die prematurely when compared to control fish. In addition, egg sizes diminished and spawning success declined, resulting in a reduction of new year-class recruitment and depressed population numbers (Franzin and McFarlane 1980). Fry are generally the most sensitive life stage of fish whereas eggs are often the most resistant to metals (Sauter et al. 1976). Copper and cadmium are of comparable toxicity to fish and an order of magnitude more toxic than lead or chromium (Sauter et al. 1976). Lead and zinc levels in benthic insects and three fish species from roadside streams were found to correlate with traffic density, yet no adverse effects were detected among fauna (Van Hassell et al. 1980).

Principle water chemistry variables influencing the availablity and toxicity of inorganic metal ions include hardness, acidity, alkalinity and rates of organic or inorganic complexation (Chakoumakos et al. 1979). Reduced water calcium levels can enhance metal uptake in aquatic fauna (Franzin and McFarlane 1980; Muniz and Lievestad 1980a). Zinc is found to be more toxic to bluegill at depressed pH levels (Cairns et al. 1972). At a given pH, less Cu is required to reduce fish growth in soft waters than hard waters; for given hardness and Cu levels, greater growth reductions are observed in waters of low than neutral pH (Waiwood and Beamish 1978). The temperature of surface waters is also found to influence metal toxicity. Salmon acclimated to cold water temperatures were observed to be significantly more resistant to Zn toxicity than warm-acclimated individuals (Hodson and Sprague 1975).

Aquatic insects are generally less sensitive than fish to metal concentrations in surface waters, however, the mayfly species and other groups are acutely sensitive to many metals (Warnick and Bell 1969; Nehring 1976). The crustaceans on which fish feed are believed to be adversely affected at metal levels that also impair fish, however daphnia are found to be more sensitive than fish (Biesinger and Christensen 1972). Effects of particulates on most zooplankton and other invertebrates largely have yet to be clarified, particularly for the case of aluminum entrainment and air pollutants such as mercury.

5.2.6 Aquatic Ecosystem Response

The potential effects of atmospheric particulates on aquatic ecosystems range widely from beneficial fertilization with nitrogen, phosphorus and potassium compounds to a variety of toxic effects from the trace metals, organic pollutants and associated acids (Galloway and Cowling 1978). However, very few ecosystem-level responses, beyond reductions in biotic density and diversity, have been documented.

Despite the lack of detailed ecological assessments, metal-impacted aquatic systems have been studied near point sources (Hutchinson and Whitby 1974; Franzin and McFarlane 1980; Keller et al. 1980), as well as in areas subject to regional deposition of pollutants transported long distances (Almer et al. 1974; Wright and Gjessing 1976; Ficke 1978; Galloway 1978; Schindler et al. 1980a; Tomlinson et al. 1980). Further impact situations have been characterized for the acid-induced mobilization of metals from sediments (Schindler et al. 1980b; Schindler 1980), for snowmelt pulses of elevated metal concentrations (Schofield 1977; Franzin and McFarlane 1980) and for chronic leaching of toxic metals from the watershed (Troutman and Peters 1980). These studies lead to the general conclusion that unusually elevated concentrations of metals are found primarily in those lakes, streams and sediments in North America and Scandinavia that receive chronic acid precipitation.

The full scope of ecosystem impact associated with freshwater acidification will be described in the following sections dealing with acidifying air pollutants. At present, the degree to which ecosystem alterations can be attributed to metals is highly uncertain. In many cases, the effects of atmospheric particulates may be entirely overshadowed either by impacts of acidification or by non-atmospheric point and non-point sources of metal and organic pollution. Trace metal depositions seldom occur in the absence of the acidifying air pollutants; thus, as discussed above, it is likely that interactions between water pH and metal toxicity determine the extent to which metals can alter ecosystem-level processes. Further research is needed to demonstrate the direct and synergistic effects of atmospheric metals on aquatic ecosystem functions.

In general, metal-induced changes of water chemistry and subsequent biotic impoverishment can be expected to cause ecosystem alterations paralleling those observed in terrestrial systems. These may include:

- increased organic litter accumulation;
- decreased decomposition and nutrient cycling;
- altered primary and secondary productivity;
- altered energy relationships between trophic levels; and
- changes in species interactions and competitive relationships.

Few, if any, of these potential impacts have been confirmed in a quantitative fashion nor linked directly with effects from atmospheric metal deposition. Although such effects have been observed in situations involving extremely high levels of contamination, such as acid mine drainage (Parsons 1968), freshwater metal levels and acidities resulting from air pollution are far less than those of acid mine drainage and the ecosystem-level impacts taking place can be expected to be more subtle and difficult to detect.

Aluminum inputs to aquatic ecosystems must continue to be studied as an important special case since this common element of the earth's crust is introduced as an undesirable by-product of acid deposition in sensitive terrestrial and aquatic ecosystems. Again, the degree of interplay between acidity and Al toxicity may determine the importance of this substance in ecosystem impacts. Further research is needed to reveal whether or not primary and secondary productivity can be reduced by Al complexation with nutrient calcium, phosphorus and magnesium, and subsequent precipitation to sediments. Also of importance will be knowledge of the long-term effects of Al pulses accompanying snowmelt on microbial activity, nutrient cycling and sustained ecosystem productivity.

5.3 RESPONSE TO ACIDIFYING AIR POLLUTANTS

The primary acidifying pollutants SO_2, NO_2, and Cl_2, in their unreacted states, generally produce acute effects on terrestrial biota only in the immediate vicinity of their release from point sources. Terrestrial plant damages from acute and chronic exposures to these air pollutants are well-documented while pollutant avoidance and other behavioral alterations have been demonstrated in terrestrial animal populations.

As described in Section 3.1 of this report, when the primary acidifying pollutants remain in the atmosphere following emission, they may be transported over long distances and undergo chemical transformations to secondary sulfates, nitrates and chlorides which contribute to both wet and dry acid deposition. In the northeastern United States, mean annual precipitation pH values are found in the range 4.0-4.5, while individual storms of pH 3.0-3.6 have been recorded (National Atmospheric Deposition Program 1978). Known effects include measurable damage to aquatic ecosystems and severe impoverishment of sensitive soils; potential long-term injuries to forests are suspected. In general, the effects of acid deposition on terrestrial plants, animals, and ecosystem-level processes are subtle and, in contrast to aquatic impacts, remain poorly understood. Despite the scientific uncertainties, acid deposition has come to be perceived as the primary environmental threat to terrestrial and aquatic systems in large portions of Scandinavia, eastern Canada and the northeastern United States (National Atmospheric Deposition Program 1978; Overrein et al. 1980; Impact Assessment Work Group 1981).

Both direct deposition, including wet and dry deposition, and surface run-off of the secondary acidifying pollutants contribute to the acidification of freshwater ecosystems. As a general rule, surface water acidification may be expected to occur in sensitive regions receiving chronic acid precipitation of pH 4.7 or less (Schindler 1979). Changes in species abundance and community composition appear at all trophic levels of the aquatic ecosystem as a result of habitat acidification and subsequent alterations of water chemistry. As pH levels drop below 5.5, many common plant, fish, and fish-food species are eliminated or reduced in numbers while a small variety of acidophilic plants and animals become more abundant (Hendrey et al. 1976). Aquatic biota may be subjected to direct physiological and reproductive stresses as well as indirect effects arising from alterations of their abiotic environment. Ecosystem functions of decomposition, mineralization and energy transfer can be disrupted by acid-induced changes in water quality and the spatial and temporal distribution of biota. The end results are often a simplification of biotic community structure, reductions of ecosystem self-regulation and stability, and arrested ecosystem succession and evolution.

With respect to aquatic and terrestrial ecosystem alike, little is known of how alterations in precipitation chemistry and atmospheric deposition can accelerate patterns of change taking place over long time periods. As with particulate accumulation in biota and other ecosystem compartments, the short-term consequences of exposures to acidifying air pollutants are often not readily apparent and the long-term potential for harm remains largely unknown. Should such long-term alterations be taking place, virtually nothing is known of their reversibility or the potential for mitigation.

Thus, continued research into the effects of acid deposition must address many fundamental questions beyond those pertaining to direct biotic effects. Entire biogeochemical cycles, in particular, should be the subject of future research; linkages between terrestrial and aquatic ecosystems are also an important concern since biotic and abiotic effects in different ecosystems may be highly interrelated (National Atmospheric Deposition Program 1978). Such studies should increase understanding of how alterations induced in ecosystem functioning and the abiotic environment can affect short-term and long-term plant and animal adaptivity, productivity, and survival, especially in relation to the top-order carnivorous birds and fish. The complex nature of questions raised by the acid deposition phenomenon tends to underline the intimate relationship between air, water, and soil contamination in the assessment of impacts to fish, wildlife, and their habitats.

5.3.1 Terrestrial Plant Response

The unreacted gaseous pollutants SO_2, NO_2, and Cl_2 enter plants via stomatal openings and exert their primary damage on the cells within leaves, creating a characteristic type of lesion for each pollutant (Jacobson and Hill 1970). Within the plant the primary pollutants are transformed to their corresponding acids by moisture in the leaf interior. Plants possess a limited capacity to reduce the contaminants to less toxic compounds, but plant injury occurs once this capacity is exceeded (Knabe 1976). Reductions in photosynthetic production, plant growth, and cellular enzyme function are subtle effects which may occur even in the absence of visible tissue damage, premature senescence, or defoliation (Ziegler 1975; Malhotra and Hocking 1976).

In general, the effects of plant exposure to acidifying air pollutants will depend upon the inherent sensitivity, stage of development, and nutritional status of the plant, as well as environmental factors governing the opening and closing of stomata and the condition of the leaf cuticle (Guderian 1977). For example, high light intensity and adequate soil moisture will increase the susceptibility of exposed plants to damage by stimulating stomatal opening and gas exchange (Glass 1979b). In many plants, particularly in locations where elemental sulfur and biologically available nitrogen are limiting, low-level exposures to primary gaseous emissions may actually enhance plant growth and productivity (Bennett et al. 1974; Knabe 1976).

Lichen species are known to be particularly sensitive to chronic low-level SO_2 pollution and a wide variety of physiological responses have been documented from laboratory and field exposures (Eversman 1978, 1980).

Table 24 lists several studies in which epiphytic lichens or their tree-bark substrates have been employed to monitor ambient levels of acidifying air pollution.

Table 24. Studies of the use of lichens and tree bark in the monitoring of acidifying air pollution.

Gilbert 1970
Le Blanc et al. 1972
Creed et al. 1973
Johnsen and Sochting 1973
Sundstrom and Hallgren 1973
Grodzinska 1976
Henderson and Seaward 1977
Skye 1979

Deposition of the secondary acidifying pollutants, or strong acids, has yet to be linked with adverse plant effects in the field. However, experimentation with simulated acid mists has suggested that terrestrial plants may be adversely affected by acid precipitation in two distinct ways (Jacobson 1980):

- by direct contact with foliage and reproductive organs, which can injure living tissues and reduce productivity, growth, yield, and forage quality; and

- by reduced soil fertility that results from gradual alterations of the physical, chemical, and biological properties of soils.

However, as noted above for low-level SO_2 pollution, chronic sulfate and nitrate deposition on plant and soil surfaces can significantly augment plant growth and production in sulfur- and nitrogen-deficient soils (Abrahamsen 1980a; Overrein *et al*. 1980; Tamm and Wiklander 1980; Tveite and Abrahamsen 1980).

The observed effects of foliar applications of simulated acid rain (usually of pH less than 4.0) include:

- abnormal cell elongation and proliferation (Evans *et al*. 1977; Evans 1980);

- erosion of leaf cuticle and epithelial waxes (Shriner 1976; Evans 1979a; Fowler *et al*. 1980);

- leaching of plant nutrients from foliage (Fairfax and Lepp 1975; Wood and Bormann 1975; Tukey 1980);

- predisposition to attack by pathogens, parasites, and insects, and inhibition of beneficial symbionts (Shriner 1976, 1978, 1980; Weber et al. 1979; Shriner and Cowling 1980);

- interference with sexual reproduction (Evans 1979b; Evans and Conway 1980; Evans and Lewins 1980); and

- necrotic lesions on foliage and reproductive structures (Wood and Bormann 1974; Irving 1978; Shriner 1978; Evans and Curry 1979; Evans 1980; Haines et al. 1980; Horntvedt et al. 1980; Lee et al. 1980).

Other direct effects postulated to occur include interference with the function of guard cells and alterations of root- and leaf-exudation processes (Tamm and Cowling 1976). The former may result in water stress and increased foliar uptake of gaseous pollutants, such as SO_2 and ozone, while the latter effect may reduce populations and activities of beneficial leaf and root symbionts such as nitrogen-fixing bacteria and fungal mycorrhizae.

Dissolved acids in rain, dew and fog are believed to collect in small pools in leaf surface depressions. Depending on the plant species, a number of protective mechanisms, summarized in Table 25, may reduce pollutant injury. Generally, however, acids are able to penetrate cracks in protective leaf cuticles near specialized structures such as veins, trichomes, and stomata (Evans et al. 1977, 1978). Necrotic lesions, pitting, chlorosis, spotting, and galls are observed most frequently near leaf veins and trichomes; injuries are found to occur at both the cellular and tissue levels, and to enlarge in a localized fashion (Shriner 1979; Evans 1980). A surficial pH of 3.4 is through to be the threshold for visible injury (Evans 1979a).

Table 25. Processes and structural characteristics that reduce plant sensitivity to acid deposition.

Exclusion	• leaf orientation and morphology • chemical composition of cuticle • flower orientation • protection of sexual organs • pollination mechanism
Neutralization	• salts on leaf surfaces • buffering capacity of leaves
Metabolic Feedback Reactions	• enzymatic reactions that consume hydrogen ions or yield alkaline products

(From Jacobson 1980)

Expanding and newly expanded leaves and needles are more sensitive to acid penetration and injury than immature and older leaves (Wood and Bormann 1974; Evans et al. 1978) while matured leaves are the most susceptible to acid-induced foliar leaching (Tukey 1980). Monocotyledons are generally thought to be less sensitive to leaf injury than dicotyledons, conifers are comparatively less sensitive than deciduous trees, and the foliage of woody, broad-leaved plants may be less sensitive than that of broad-leaved, herbaceous plants (Evans 1980).

Plant effects caused by reduced soil fertility and increased acidity are not as well understood as direct foliar impacts. While reductions in available phosphorous have not been documented, accelerated soil leaching of nutrient calcium, magnesium, potassium, iron and manganese is known to occur (Berigari and Xerikos 1975; Reuss 1978, 1980; McFee 1979).

In soils of low pH, seedling establishment has been shown to be more sensitive to acidity than the germination process (Lee and Weber 1979). Seedling growth has been found to be enhanced in some tree species and reduced in others (Wood and Bormann 1974, 1977). Field investigations of forest tree growth have yet to provide evidence of adverse effects in areas receiving acid precipitation (Cogbill 1976; Overrein et al. 1980). Long-term research and improved research methods will be required to adequately assess potential acid-induced impacts to native plant species in their natural environments.

5.3.2 Terrestrial Animal Response

The effects of acidifying air pollutants on terrestrial vertebrates are expected to be primarily indirect, involving alterations of soils and vegetation which, in turn, may lead to modifications of food resources and the habitats of animal communities (Dvorak et al. 1978). For example, arctic caribou are dependent on lichens as their sole source of nourishment, yet these are known to be among the most sensitive of terrestrial plant species to low-level SO_2 exposures (Schofield et al. 1970). Much more research is needed to link the nutritional requirements and feeding habits of wildlife species with the effects of the acidifying pollutants on plants.

Information on the direct effects of sulfur and nitrogen oxides has been derived almost exclusively from toxicological experiments conducted in the laboratory (Lillie 1970; Kavet and Brain 1974). These studies generally do not provide well-defined exposure thresholds for physiological injury or death, nor are they conducted at the low exposure levels most commonly encountered in the field (Dvorak et al. 1978). On the other hand, terrestrial wildlife populations are seldom found in areas with high ambient concentrations of sulfur dioxide or sulfates. Acute effects on animals are thus restricted to areas very near point sources of the acidifying air pollutants. The most prevalent effects of these pollutants are irritations of the eyes and respiratory tract (Newman 1975, 1980) which can lead to emigration, a variety of behavioral alterations or reductions in inter- and intra-specific competitiveness (Chilgren 1978).

Laboratory studies undertaken using mixtures of air pollutants have shown that there is often a synergistic or antagonistic effect between sulfurous pollutants and other components of the atmosphere. Amdur (1975) has shown that the bronchial irritant effect of SO_2 is enhanced by the presence of sodium chloride aerosols. Substances such as potassium chloride, ammonium sulfate, ferrous oxide, magnesium and vanadium were all shown to act synergistically with SO_2 in increasing lung airway resistance. Materials with buffering or reducing capacities appeared to have a protective effect in opposition to SO_2 and thereby prevented or minimized observed reductions in pulmonary function.

Field work conducted in Czechoslovakia indicates that a variety of other direct effects may occur in vertebrate populations near point sources of SO_2 and particulate pollution. Changes in reproductive coefficients as well as physiological alterations of blood calcium and protein levels, hematocrit and hemoglobin, and urine chemistry have been documented in rabbit populations exposed to mixed industrial emissions (Novakova 1969, 1970; Novakova and Roubal 1971; Novakova et al. 1973). The potential for these and other subtle impacts on animals remains to be investigated in North America.

No direct impacts on terrestrial wildlife populations have been documented from exposures to acid precipitation. Although research on the indirect effects of diminished food supply and habitat degradation due to acidification is just beginning, these effects are thought to be of considerable importance. Table 26 provides a list of some predominantly terrestrial species which may be subject to indirect effects from aquatic ecosystem acidification. Other species which may be impacted in a like fashion include eagles, ospreys and other avian piscivores, as well as bears, gulls, and other species that regularly exploit the annual salmon runs in North American rivers and streams.

In the Adirondack Mountains of New York and the LaCloche Mountains of Ontario, common loons continue to nest and fish at lakes that no longer sustain viable fish populations (Harvey 1979; Trivelpiece et al. 1979). Fully seventy percent of common loon and hooded merganser populations inhabit areas receiving acid deposition while approximately half of the range of the bald eagle is similarly threatened (Peakall 1979). Almer et al. (1978) have suggested that many piscivorous birds may be forced to migrate from large areas impacted by acid deposition. The potential magnitude and permanence of these wildlife effects will depend on the severity of aquatic animal responses to freshwater acidification, as discussed below in Section 5.3.5.

Table 26. Birds and mammals susceptible to indirect effects of acid deposition.

Common loon (*Gavia immer*)	Mallard (*Anas platyrhynchos*)
Common merganser (*Mergus merganser*)	Black duck (*Anas rubripes*)
Hooded merganser (*Mergus cucullatus*)	Green-winged teal (*Anas carolinensis*)
Red-breasted merganser (*Mergus serrator*)	Northern pintail (*Anas acuta*)
Great blue heron (*Ardea herodias*)	American widgeon (*Anas americana*)
Belted kingfisher (*Megaceryle alcyon*)	American mink (*Mustela vison*)
Common goldeneye (*Bucephalus clangula*)	River otter (*Lontra canadensis*)
Ring-necked duck (*Aythya collaris*)	Muskrat (*Ondatra zibethicus*)

(Adapted from Impact Assessment Work Group 1981)

5.3.3 Terrestrial Ecosystem Response

The dominant ecosystem-level effects of the acidifying air pollutants and acid precipitation involve alterations of soil chemistry, fertility and structure, which can diminish the productivity of habitat. As discussed above in relation to terrestrial plant responses, cations essential to plant growth such as calcium, magnesium and potassium may be subject to accelerated leaching from terrestrial ecosystems as a result of soil acidification (Abrahamsen 1980a, b; Galloway *et al.* 1980; Wright and Johannessen 1980). As soils become more acidic, the biological availability of phytotoxic metals may increase (Overrein *et al.* 1980). Abrahamsen (1979) studied the relation of acid deposition to productivity in terrestrial ecosystems and identified three important factors related to soil processes which require study:

- the function of bicarbonate and aluminum systems in soils;
- the efficiency of cation exchanges with hydrogen ions; and
- comparisons of anion inputs in precipitation with leaching outputs to the catchment.

In discussing the fertilization effect of nitrate and sulfate deposition in terrestrial ecosystems, Abrahamson (1980a) has warned that the long-term effects of nitrate and sulfate enrichment may be comparable to those of incomplete fertilization: plant growth may be stimulated by nitrogen and sulfur inputs, while deficiencies of potassium, magnesium, phosphorus and other micronutrients are simultaneously reinforced. Studies of anion budgets in well-buffered watersheds of Tennessee suggest that the majority of deposited sulfur is retained within soils, while approximately 20 to 30 percent is drained into aquatic systems (Kelley 1980). However, in young, poorly buffered soils of low cation exchange capacity, sulfate adsorption in soils may be considerably reduced and atmospheric sulfate inputs may equal drainage losses (Kerekes 1980; Skartveit 1980; Wright and Johannessen 1980). Nitrates, on the other hand, are largely retained in both well and poorly buffered watersheds due to rapid uptake and utilization by flora. Thus, sulfate mobility will largely influence rates of soil cation leaching and can produce the equivalent of an oligotrophication effect, while nitrates will have a negligible effect.

Ecosystem productivity may also be adversely affected by acid-induced changes in the structure and activity of soil microfloral communities. Such alterations, due to increased soil acidity, have been documented for individual populations of bacteria, fungi, and algae (Bryant et al. 1979; Alexander 1980a,b; Baath et al. 1980; Lohm 1980). Microbial processes consequently affected may include (Alexander 1980a,b; Francis et al. 1980):

- contributions to soil structure, such as soil aeration and the biosynthesis of humus;
- the detoxification of aromatic substances and other phytotoxic metabolites derived from incomplete decomposition;
- the detoxification of accumulated anthropogenic pollutants; and
- reactions fundamental to sustained plant growth, including the decomposition of organic matter, the mineralization of organic phosphorus and other nutrients, ammonification, nitrification, and nitrogen fixation by blue-green algae and plant symbionts.

Macrofaunal responses are also important to ecosystem productivity as these organisms aid in decomposition processes and contribute to humus conditioning and to the maintenance of soil structure (Voigt 1980). Reductions in species numbers and avoidance behavior have been shown to result from SO_2 fumigation in a variety of insects ranging from decomposer beetles to grasshoppers (Bromenshenk 1979, 1980; Leetham et al. 1980a,b; McNary et al. 1980). Population fluctuations have been elicited by experimental soil acidification with artificial acid rain; the abundance of springtails (Collembola) in particular diminishes with increasing soil acidity (Hagvar 1980; Overrein et al. 1980). These studies raised the possibility that reproductive success in soil macrofauna is primarily a function of soil pH.

Food chain effects, similar to those observed with particulate pollution, have not been specifically associated with exposures to acidifying air pollutants. Nevertheless, reductions in primary productivity may have repercussions on secondary productivity through a diminished carrying capacity of habitat. Moreover, nutrient exports from aquatic to terrestrial ecosystems may decline significantly in response to effects on waterfowl and other piscivorous species noted in the previous section. Such exports are important insofar as they comprise one of the primary links by which nutrient matter drained into surface waters is returned for recycling in terrestrial ecosystems.

5.3.4 Aquatic Plant Response

Freshwater acidification has been shown to cause drastic alterations of all major aquatic floral groups. Altered water chemistry can directly impair some species, notably phytoplankton, although toxicity mechanisms and the individual effects of excess hydrogen, sulfate and metal ions remain to be clarified. Acidophilic species, including some plankton, mosses, and filamentous algae, increase in abundance under acidified conditions. In general, the nature of biotic impacts produced through modifications of the abiotic environment determines the extent of indirect effects on aquatic flora. Alterations of aquatic macrophyte populations, for example, often occur in response to changes in species composition and other aspects of ecosystem structure and function induced by direct impacts on biota.

The phytoplankton typically respond to acidic conditions by increasing or decreasing in numbers, species diversity, and proportion of community biomass. Populations of many species decline or vanish while those of a few proliferate, resulting in an overall decrease in the diversity of taxa present. The exact nature of shifts in species numbers and community structure remain obscured by conflicting observations in acidifying environments of differing locations (Kelso et al. 1981; Schindler 1980).

Moss (1973) investigated the pH tolerances of freshwater phytoplankton and found the lower growth limits for most species to lie between pH 4.5 and 5.1; however, as pH drops, the acidophilic Euglena sp. (a flagellate form) and Eunotia sp. (a diatom) survive to pH levels of 3.65 and 3.9, respectively. Eunotia exigua, in particular, has been found to be one of the most acid-tolerant species of diatoms (Van Dam et al. 1980). Figure 22 presents average minimum pH tolerances of major phytoplankton groups inhabiting freshwaters of North America. These averages must be used with caution, however, since responses of individual species may vary

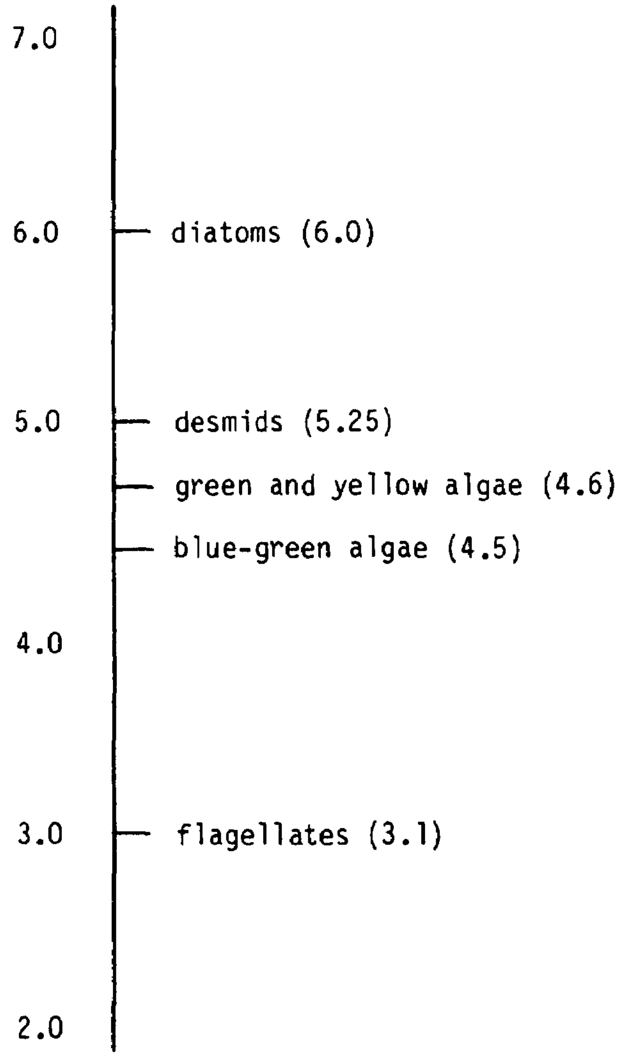


Figure 22. Lower limits of pH tolerance among the phytoplankton. (Adapted from Eilers and Berg 1981)

greatly, as noted above for diatoms. Virtually all of these groups contain some acidophilic species (Eilers and Berg 1981).

In Scandinavian lakes, the most rapid reductions in species numbers of phytoplankton were found to occur at pH 5.5 (Leivestad et al. 1976) and most population shifts took place within the 5.0 to 6.0 pH interval (Almer et al. 1974). This finding was supported by Kwiatkowski and Roff (1976) who reported that diversity indices of phytoplankton communities in the La Cloche Mountain lakes of Ontario declined significantly as water pH fell below 5.6; the following reductions in species diversity were observed:

- green algae (Chlorophyta) were reduced from 26 to 5 species;
- yellow algae and diatoms (Chrysophyta) decreased from 22 to 5 species; and
- blue-green algae (Cyanophyta) declined from 22 to 10 species.

Blue-green algae were observed to predominate over green algae in these acidified lakes, contradicting observations in Scandinavia (Almer et al. 1974) and Florida (Crisman et al. 1980) where blue-green algae were found to decline in acid lakes. Also in southern Ontario, populations of the flagellate Dinophyceae were found to increase in response to declining pH, largely replacing the diatoms and Chrysophyceae; when present as a significant fraction of producer biomass, dinoflagellates have been suggested as a reliable indicator of freshwater acidification (Yan 1978; Yan and Stokes 1978; Yan 1979). Despite the conflicting reports on responses of many phytoplankton species, there is general agreement that species numbers of green algae and diatoms decrease in acid lakes (Conroy et al. 1976; Almer et al. 1978).

Reductions in numbers of phytoplankton taxa also have been recorded in Adirondack Mountain lakes of decreasing pH (Hendrey 1980). Figure 23 shows the number of taxa observed in three lakes of varying pH. Crisman et al. (1980) reported an average of 10.8 species in acid lakes of central Florida while non-acid lakes averaged 16.5 species; phytoplankton exhibited greater population declines than members of any other trophic level in these lakes. It is generally difficult to attribute effects on these populations to excess hydrogen ions alone, as metal toxicity or nutrient deficiencies may also play an important role (Hendrey 1979; Hendrey et al. 1980c).

Observations of the effect of acidification on overall biomass and productivity are contradictory. In some acid lakes, phytoplankton biomass and productivity have shown an overall decline (Hendrey et al. 1976; Hendrey 1978). In Florida, Crisman et al. (1980) recorded mean annual chlorophyll concentrations of 1.88 mg/m^3 in acid lakes, whereas non-acid lakes possessed 7.53 mg/m^3; phytoplankton numbers were 5,700/ml and 14,000/ml respectively. In other acid lakes, these parameters are found to remain comparable with surrounding non-acid, oligotrophic lakes (Yan

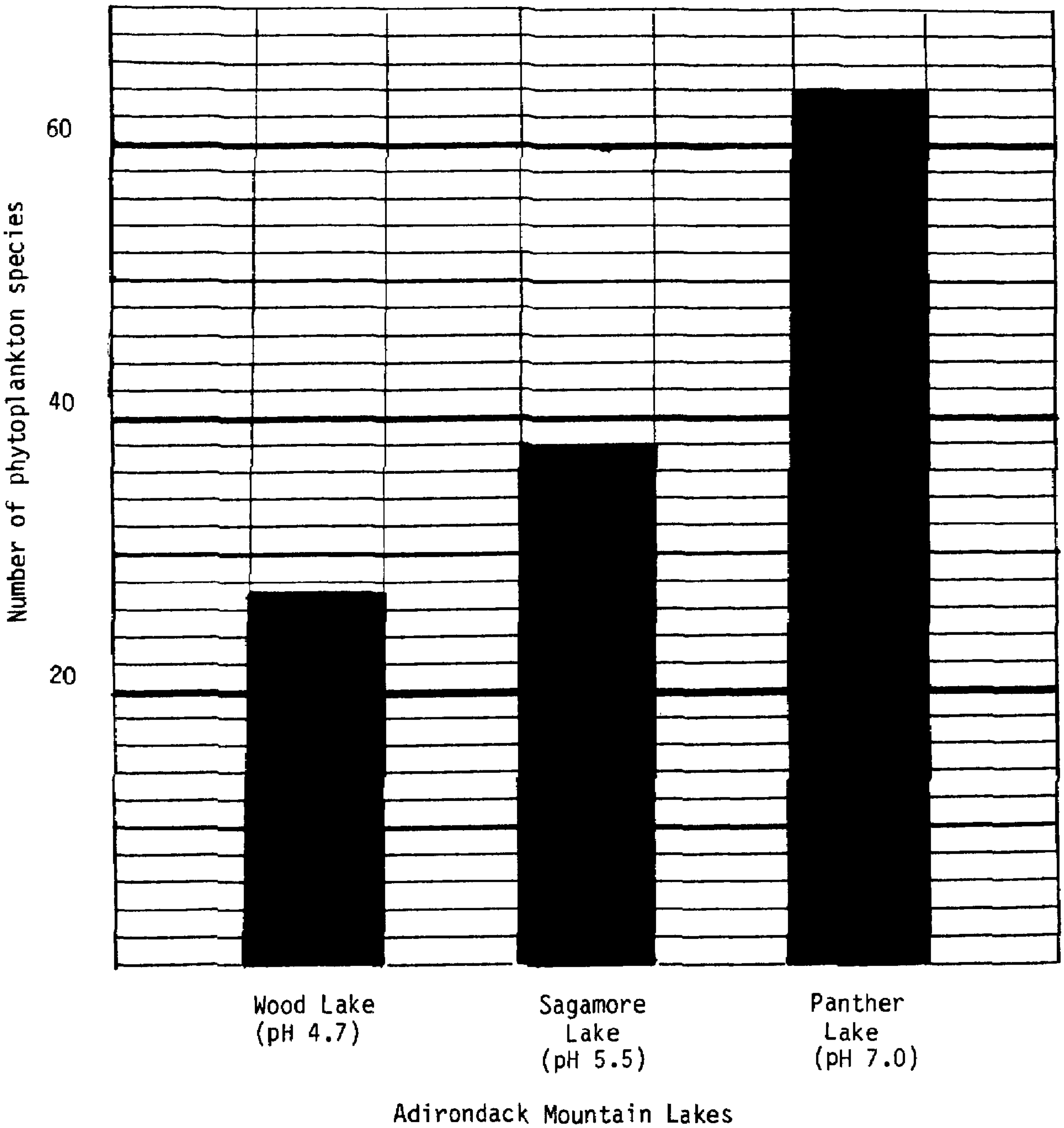


Figure 23. Number of phytoplankton species observed in Adirondack Mountain lakes of different pH. (From Hendrey 1979)

1979), while in still others, phytoplankton biomass is observed to increase (Malley et al. 1981). In acid lakes of New York, yearly increases in phytoplankton biomass and productivity are found to be atypical of those in nearby non-acidic lakes as they occur much later in the season following spring thaw (Hendrey 1980).

The decline of phytoplankton productivity and biomass per unit of water volume may be accompanied by an increase in biomass and productivity per unit area. This happens because eliminations and reductions of some species leads to increased water transparency, deepening the euphotic zone. The remaining populations expand to deeper water strata (Almer et al. 1978). Kwiatkowski and Roff (1976) observed that high levels of productivity on a volumetric basis were maintained above pH 4.8, yet subsequent reductions in standing crop were offset by the increased transparency and elevated areal production was maintained to pH 4.5. Thus, in many cases, no net loss or gain will accompany increased hydrogen ion loadings (Yan 1979; Hendrey et al. 1980c). Phytoplankton biomass and productivity are therefore less reliable as an indicator of freshwater acidification than changes in species abundance and composition (Yan and Stokes 1978).

The recent pH history of lakes for which water quality data are not available can be inferred from stratigraphic studies of the diatom composition of surface sediments (Almer et al. 1978; Singer 1981). The percentage of acidophilic diatom species present in sediments of Norwegian lakes was found to be inversely correlated with increasing depth of the sample, thus indicating the time frame over which lake acidification had progressed (Davis and Berge 1980).

Responses of mosses and benthic filamentous algae to freshwater acidification are generally very similar. Both of these plant groups typically increase in numbers and proportion of community biomass. In the absence of aquatic acidification, benthic algae are usually found in small numbers while the mosses, which are primarily terrestrial or semi-aquatic species, may be absent or confined to shallow littoral areas. The increased water transparency accompanying freshwater acidification permits them to inhabit depths where sufficient light would not ordinarily be available. However, the mechanisms which enhance the competitiveness of these species are not yet clearly understood.

Both the mosses, primarily *Sphagnum* and *Fontinalis* species, and filamentous *Mougeotia*, *Batrachospermum* and *Zygogonium* species form heavy mats in association with fungi and organic matter. These mats effectively seal profundal sediments (Grahn et al. 1974; Hultberg and Grahn 1976; Hultberg 1978). The invasion of *Sphagnum* species has been observed to begin at pH 6.0 and intensify greatly below pH 5.0 (Hendrey and Vertucci 1980); it is found to be greatest in shaded portions of the littoral zone (Grahn 1977). The spread of filamentous algae first occurred at pH 5.5 in experimentally acidified shield lakes of Canada (Schindler 1979). Hall and Likens (1980) reported periphyton biomass to increase to 1.7 ug chlorophyll a/cm^2 in acidified streams (pH 4.0) from a value of 0.3 ug chlorophyll a/cm^2 in a reference stream (pH 6.4). Dense growths of filamentous algae have also been reported in lakes of low pH near Sudbury, Ontario (Keller *et al.* 1980). Both mosses and periphytic algae are abundant in acid streams of Scotland (Harriman and Morrison 1980), in the lakes of the Adirondacks (Hendrey and Vertucci 1980), and in Scandinavian lakes (Hultberg and Grahn 1976; Almer *et al.* 1978; Overrein *et al.* 1980).

There is little evidence that aquatic vascular plants are directly affected by freshwater acidification, at least in the absence of heavy metals (Gorham and Gordon 1963), although reduced growth and flowering activity in *Lobelia dortmanna* is a documented response to lowered water pH (Leivestad *et al.* 1976). Indirect effects, however, can be marked as submergent macrophytes are displaced by expanding populations of aquatic mosses and algae that shade necessary light, deplete nutrients and render stream and lake bottoms unfavorable for the growth of these species (Grahn 1977). In acid Lake Colden of New York, *Lobelia*, *Littorella*, and *Isoetes* species were driven out by invasions of mosses, primarily *Sphagnum pylaesii*; other decreasers included *Nuphar leuteum* and *Eleocharis acicularis* while numbers of *Utricularia* and *Eriocaulon* species typically increased.

Some tolerant species may colonize suitable substrates at greater depths. *Juncus* species are acidophilic and also increase in response to elevated accumulations of organic matter (Hultberg and Grahn 1976). Furthermore, the benthic production of macrophytes may be comparatively greater in acid than non-acid lakes as it is for invading mosses and filamentous algae (Almer *et al.* 1978). Nonetheless, the end result will be an overall simplification of the macrophyte community (Singer 1981).

Responses of aquatic flora to acidification may be summarized as follows (Burton *et al.* 1981):

- species shifts to acid-tolerant forms;
- lowered species diversity; and
- increased standing crops of some species concurrent with decreased standing crops of others.

Changes in primary productivity cannot be predicted.

5.3.5 Aquatic Animal Response

Both vertebrate and invertebrate forms of aquatic fauna are susceptible to adverse impact from freshwater acidification. Direct and indirect effects on fish and fish-food organisms occur at all trophic levels. Common responses involve alterations of the osmoregulatory processes freshwater organisms use to maintain the ionic strength of internal tissue fluids, leading to subsequent changes in cell metabolism and energy use. Indirect effects are also common in impacted aquatic ecosystems and occur in response to modified plant and animal populations or other acid-induced alterations of habitat.

Communities of zooplankton and zoobenthos characteristically respond to freshwater acidification by diminishing in species diversity and abundance. Fewer taxa and lower mean numbers of zooplankton and benthic invertebrates, compared with control lakes, have been documented in acid lakes of Sweden (Almer et al. 1974), Norway (Hendrey and Wright 1976; Hendrey et al. 1976; Leivestad et al. 1976; Raddum 1980), Canada (Sprules 1975; Conroy et al. 1976; Roff and Kwiatkowski 1977; Yan and Strus 1980; Keller et al. 1980), and Florida (Crisman et al. 1980). The number of dominant species in the zooplankton community is found to be less in acidified waters (Keller et al. 1980); Sprules (1975) noted only one or two dominants of 1-7 species present in acid lakes compared with 3 or 4 dominants out of 9-16 species in non-acid lakes. Factors affecting invertebrate responses to acidification are thought to be very complex, however, altered dominance patterns in acid lakes are caused primarily by (Overrein et al. 1980):

- differences in the physiological tolerance of individual species (direct effects); and

- the absence of predatory fish populations (indirect effects).

Some of the most sensitive fish-food organisms, including the tadpole shrimp (Lepidurus arcticus) and common gammarus (Gammarus lacustris), are eliminated below pH 6.0 in Scandinavian lakes (Borgstrom and Hendrey 1976; Hendrey 1979). The opossum shrimp, Mysis relicta, and other crustacean fish-food organisms of North American lakes are reported to be equally sensitive (Schindler 1979; Malley et al. 1981). Reductions of these populations may serve as an early warning of ecosystem acidification.

Acidity tolerances of species representative of the major groups of fish-food organisms are presented in Figure 24. An extensive review of invertebrate tolerances to pH has been prepared by Eilers and Berg (1981). Like the gammarus, molluscs are consistently shown to be absent from waters of pH below 6.0 (Okland 1969; Okland, J. 1980; Okland, K. 1980; Okland and Okland 1980). The freshwater louse Asellus aquaticus is more tolerant of intermediate pH levels; its disappearance from a lake signifies that fish populations are likely to be impacted. Populations of the mayfly, Baetis rhodanis, an important food-chain link in oligotrophic lakes of Norway, begin to disappear at pH levels below 5.0 (Overrein et al. 1980). Oligochaete worms may be inhibited by metal accumulations in the sediments of acid lakes (Raddum 1980).

Calcium availability is thought to play a major role in the responses of invertebrates, especially shell organisms and moulting crustaceans, to acidification (Singer 1981). In a stream experimentally acidified to pH 4.0, Burton et al. (1981) found populations of the isopod Asellus intermedius reduced to ten percent of their original numbers while the snail Physa disappeared entirely. Decreased emergence of aquatic insects, particularly midges (Diptera) and mayflies (Ephemeroptera), have also been observed in laboratory and field experiments (Bell 1970, 1971; Fiance

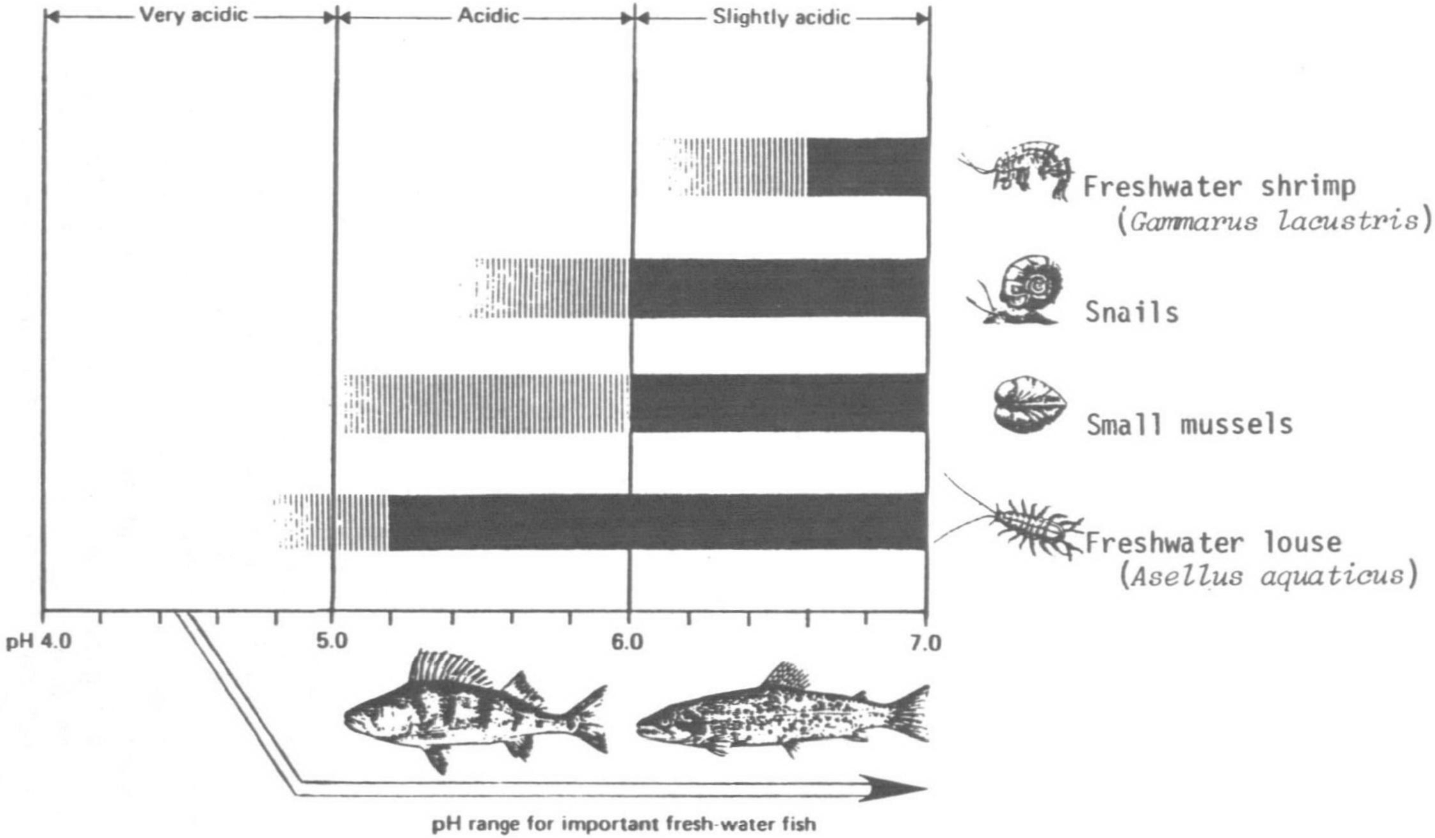


Figure 24. Comparative pH tolerances of four groups of fish-food organisms. (From Okland and Okland 1980)

1978). Lowered densities and increased drift have been observed in benthic insects of acidified streams (Hall *et al.* 1980; Hall and Likens 1980).

Some invertebrates, notably the shredder organisms (*Trichoptera*), are acidophilic and are found to assume a greater proportion of total benthic biomass in impacted ecosystems (Friberg *et al.* 1980; Burton *et al.* 1981). The waterboatman (*Corixidae*) and other surface-dwelling insects of stream pools and lakes are particularly tolerant of low pH and may proliferate in the absence of fish predators (Rosseland *et al.* 1980). Nevertheless, the caloric value of these fish-food organisms has been found to decline in acidified waters as invertebrates in general must devote more energy to the maintenance of osmoregulatory balances (Hendrey 1979; Hall and Likens 1980). A detailed review of benthic invertebrate responses has been prepared by Singer (1981).

Amphibians are an essential food-chain link as predators of invertebrates and as prey for fish and waterfowl. Amphibian populations may be particularly sensitive to acid deposition if they breed in temporary vernal pools following snowmelt. Furthermore, the range of many North American amphibian species is found to overlap areas receiving acid precipitation (Peakall 1979).

Some amphibious species which are susceptible to reduced reproductive success resulting from habitat acidification are listed in Table 27. Pough (1976, 1978) has demonstrated reductions of up to 90 percent in the hatching success of the spotted salamander (*Ambystoma maculatum*) and Jefferson salamander (*Ambystoma jeffersonianum*) at pH levels below 6.0. Larvae and young of the shovel-nosed salamander (*Leurognathus marmoratus*) have been shown to be more sensitive to acidic conditions than adults of the species (Mathews and Larson 1980). Reproduction in frogs has also been shown to be adversely impacted by depressed water pH (Gosner and Black 1957; Overrein *et al.* 1980).

The effects of acidifying air pollutants on fish populations are generally well-documented and have been reviewed by Howells and Holden (1979), Fritz (1980) and Fromm (1980). Observed reductions in fish populations have been linked to lake acidity in Norway (Leivestad *et al.* 1976; Muniz and Leivestad 1980a), Sweden (Almer *et al.* 1974; Dickson 1975), the Adirondacks (Schofield 1976) and Ontario (Beamish and Harvey 1972; Harvey 1975, 1980). Sudden fishkills have been observed following rapid drops in pH resulting from heavy autumn rains and spring snowmelt in rivers and streams of New York (Schofield 1977) and Scandinavia (Jensen and Snekvik 1972; Leivestad and Muniz 1976; Hultberg 1977; Wright and Snekvik 1978). The wide range of effects observed in individual fish, whole populations, and communities are summarized in Table 28. However, the mechanisms of acid-induced, physiological alteration taking place in the field are poorly understood. Physiological stress, periodic mass mortality, and reproductive impairment, as discussed below, all contribute to recruitment failure and the extinction of fish species in areas sensitive to acid deposition (Schofield 1980).

Table 27. Amphibians susceptible to reduced reproductive success from breeding-habitat acidification.

Species	Breeding Habitat
Rana catesbiana (bullfrog)	Lakes
Eurycea bislineata (northern two-lined salamander)	Streams
Ambystoma maculatum (yellow-spotted salamander) Ambystoma laterale (blue-spotted salamander) Bufo americanus (American toad) Rana sylvatica (wood frog) Pseudacris triseriata (chorus frog) Hyla crucifer (northern spring peeper)	Temporary meltwater ponds
Notophthalmus viridescens (red-spotted newt) Necturus maculosus (mud puppy) Rana clamitans (green frog) Rana pipiens (northern leopard frog) Rana septentrionalis (mink frog) Hyla versicolor (gray tree frog)	Permanent ponds

(Adapted from Impact Assessment Work Group 1981)

Table 28. A summary of effects of freshwater acidification on fish.

ORGANISMS	POPULATIONS
Acute mortality	Altered density
Ionic balance stress	Altered birth rates and death rates
Reduced blood pH	Altered age pyramid
Suffocation or anoxia	Extinction
Gill damage	
Spinal deformation	**COMMUNITIES**
Histological changes	
Disrupted calcium metabolism	Reduced population abundance
Trace element bioconcentration	Reduced species diversity
Reduced growth rate	Diminished autotrophic and heterotrophic production
Altered feeding behavior	Reduced biomass
Decreased resistance to environmental stress	Species replacements
Disrupted reproductive behavior	Species eliminations
Failure to reach spawning condition	Altered inter- and intra-specific relations
Delayed or reduced spawning	
Delayed egg hatching	
Reduced egg viability	
Reduced survival of life stages	

The failure of fish to successfully regulate body salt content is believed to be the primary cause of fish mortality in acidified waters. Altered sodium flux is caused by changes in the ion permeability of fish gills; active salt uptake mechanisms are inhibited by low pH, resulting in a reduced salt content of blood and body tissues (Leivestad et al. 1976; McWilliams and Potts 1978; Muniz and Leivestad 1980a, b). A rapid, preferential influx of hydrogen ions can cause acidosis in fish blood which is presumed to interfere with a variety of physiological mechanisms (Fromm 1980). Accompanying symptoms often include gill clogging with mucous, hyperventilation, increased respiratory rates, and lowered blood oxygen tension (Overrein et al. 1980; Rosseland 1980).

Research into the effects of acid precipitation on fish populations has pointed to aluminum toxicity as a major cause of injury and death to aquatic fauna in impacted environments (Cronan and Shofield 1979). This hypothesis was proposed when initial studies of freshwater acidification revealed certain responses that were difficult to explain (Schofield 1976):

- fish mortalities in natural waters were observed to occur at pH levels shown not to be acutely toxic in the laboratory; and

- the impact on fish populations was found to vary considerably in lakes of comparable acidity.

It became apparent that one or several additional toxic agents operate synergistically with freshwater acidity to impair aquatic fauna, especially during the spring snowmelt. Elevated concentrations of aluminum have since been discovered and documented in acid lakes of Scandinavia and North America (Wright and Gjessing 1976; Beamish and Van Loon 1977; Henriksen and Wright 1978; Galloway and Likens 1979), in groundwater (Grahn 1980), in the snowpack (Schofield 1977; Seip 1980) and in soils sensitive to acid precipitation (Hall et al. 1980; Hermann and Baron 1980). Water concentrations of aluminum have been found to correlate with freshwater pH (Johnson, N. 1979; Cronan and Shofield 1979; Wright et al. 1980). Thus aluminum toxicity is under extensive investigation as a major cause of fish mortality in waters of low pH (Baker and Schofield 1980; Muniz and Leivestad 1980a, b; Schofield and Trojnar 1980).

Gill destruction has been shown to be the primary biotic effect of aluminum on fish (Muniz and Leivestad 1980a, b; Schofield and Trojnar 1980). At the time of snowmelt, aluminum toxicity may be sufficiently severe to cause anoxia and mortality in fish populations (Schofield 1980). All of these effects are found to occur at water pH values (4.4-5.2) that of themselves would not produce gill damage or any other major physiological stresses in fish; in fact, elevated aluminum concentrations may exert an antagonistic effect on acid stresses to fish at pH values below 4.4 (Leivestad et al. 1980; Schofield 1980; Schofield and Trojnar 1980).

Experiments with brook trout showed that while sensitivity to low pH decreases with increasing age of the individual, sensitivity to elevated aluminum levels increases with increasing age; fry are the most sensitive life stage to combined low pH and elevated aluminum while the effects of low pH on eggs are largely mitigated by increasing aluminum concentrations (Baker and Schofield 1980). In sum, the distinction of various forms of aluminum present in water, as well as the timing of aluminum and hydrogen ion pulses in relation to the presence of sensitive fish life-history stages, are important considerations in the evaluation of potential aluminum effects on indigenous fish populations.

Acid-induced physiological stress can lead to diminished fish growth and a variety of other sublethal impacts (Beamish et al. 1975; Ryan and Harvey 1980). Physical deformities observed in fathead minnows (Mount 1973) and white suckers (Beamish 1972; Beamish et al. 1975) may be due to calcium resorption from bone tissue (Harvey 1979). Both epithelial cell degradation and altered blood protein function can predispose fish to infection or disease (Daye and Garside 1976; Falk and Dunson 1977; Fromm 1980). A generally weakened condition may diminish feeding efficiency and other competitive advantages while increasing the likelihood of predation.

Fish tolerance to depressed water pH is found to increase in waters of high conductivity and plentiful calcium concentration (Howells and Holden 1979; Muniz and Leivestad 1980a; Leivestad et al. 1980). At the same time, naturally acidified waters are more toxic to adult fish and salmonid eggs, alevins, and fingerlings than water experimentally acidified with sulfuric acid, possibly due to elevated trace element levels. Other factors modifying fish responses to acidification include (Howells and Holden 1979):

- age, size, sex, condition;
- sexual maturity;
- acclimation;
- genetic tolerance;
- ambient temperature; and
- the duration of acid events.

Seasonal changes are also an important factor as overwintering fish may be in their weakest condition during spring snowmelt, or fish may be subjected to additive thermal stresses in summer (Fritz 1980). The existence of a genetic basis for pH tolerance and survival has been established for brown trout, and the role of genetic variation in salmonid population responses is a subject of intensive research (Gjedrem 1976, 1980; Edwards and Gjedrem 1979).

Reproductive failure in fish populations often results from an inability of the female to reach spawning condition, either through altered breeding behavior or disruptions of endocrine functions. Abnormal serum calcium levels, observed in white suckers that failed to spawn in acidified waters, have been linked to the latter mechanism (Beamish et al. 1975). Other reproductive effects involve the tolerance of fish eggs and developing embryos. The genetic material of developing fish ova is susceptible to acid damage and the maturation of ova in several species is prevented by acid conditions (Menendez 1976; Ruby et al. 1977; Kennedy 1980). Schindler (1979) reported an elevated incidence of embryonic mortality and deformity in fathead minnows at pH 5.8, despite the fact that adults survived pH values above 5.5. The failure of population recruitment, in spite of spawning, results from the increased sensitivity of fish eggs, fry, and fingerlings, and may explain a major portion of observed population reductions (Rosseland et al. 1980). Perch, trout, and white suckers are all found to vary in pH sensitivity at different stages of their life history (Beamish et al. 1975; Harvey 1979).

The spawning of mature fish may also be disrupted by acid-induced invasions of benthic flora that alter the suitability of breeding sites

(Fritz 1980). Stream acidification is known to provoke avoidance reactions in brown trout populations, possibly forcing them from preferred nesting areas (Hall et al. 1980). As shown in Figure 25, this species tends to gather in areas of most favorable water quality during pronounced acidification events. Fish are particularly vulnerable to physiological stress during spring when ice cover and depleted oxygen in deep waters trap populations in surface layers while the early snowmelt is increasing the acidity of the water (Schofield 1980). The variability observed in fish population responses to lake acidity may in large part be accounted for by differences in the availability of areas protected from acidification (Muniz and Leivestad 1980a).

In Norway, fish population losses first observed in the 1940s were found to accelerate after 1960. As shown in Figure 26, an increasing percentage of lakes in four size classes have lost their brown trout population over time. Rates of population decline are greatest in southern Norway; trends in fish population status indicate that 80 percent of all trout will be lost by 1990 as a result of habitat acidification (Muniz and Leivestad 1980a). Sixty percent of brown trout populations above 800 meters in Norway are now extinct and trout populations are absent from over 200 high-altitude lakes of New York and Ontario where they once were common (Harvey 1979). Species of lake trout, brown bullhead, white sucker and several cyprinids have been eliminated from Adirondack lakes (Schofield 1976). In streams and rivers, brook trout populations are thought to be the species most impacted by acid deposition in the Adirondacks (Pfeiffer and Festa 1980) and Great Smokey Mountain National Park (Mathews and Larson 1980).

Failure in the recruitment of new age classes, due to physiological and reproductive stress, is known to be the chief mechanism of fish population extinction (Jensen and Snekvik 1972; Schofield 1976; Ryan and Harvey 1977). Accompanying losses of older fish are also common, resulting in large-scale alterations of formerly stable age distributions (Beamish et al. 1975; Harvey 1980; Rosseland et al. 1980). Severe snowmelt effects in any given year may contribute to the selective absence of different year classes in acidified surface waters (Hultberg 1977). Recruitment failure may also lead to gene pool reductions which limit potential selection for genetic tolerance in surviving populations (Beamish and Harvey 1972). Indirect effects can result from altered inter- and intra-specific competition (Henriksen et al. 1980). For example, the relatively acid-tolerant rock bass has been observed to increase in size due to reduced competition for available food (Ryan and Harvey 1977), while piscivorous fish may face a loss of juvenile prey and resort to cannibalism of their own young (Fritz 1980).

Many questions remain about the exact nature and extent of acid deposition effects on aquatic fauna. Establishing dose-response relationships for organisms is complicated by the need to control for other factors, such as trace element concentrations and the duration of acidic episodes (Fritz 1980). Additional research is needed to correlate fish population

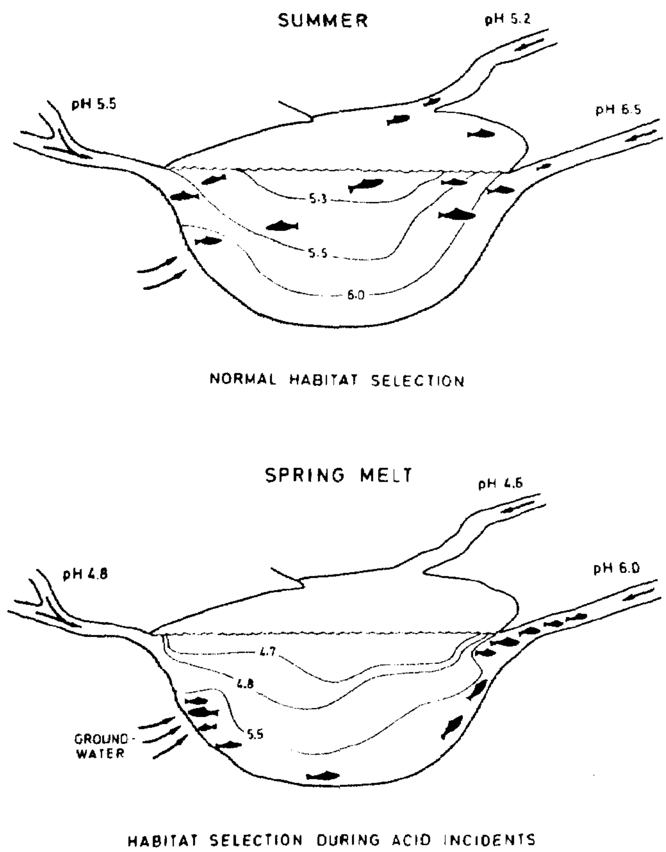


Figure 25. Fish habitat selection in response to freshwater acidification. (From Muniz and Lievestad 1980a)

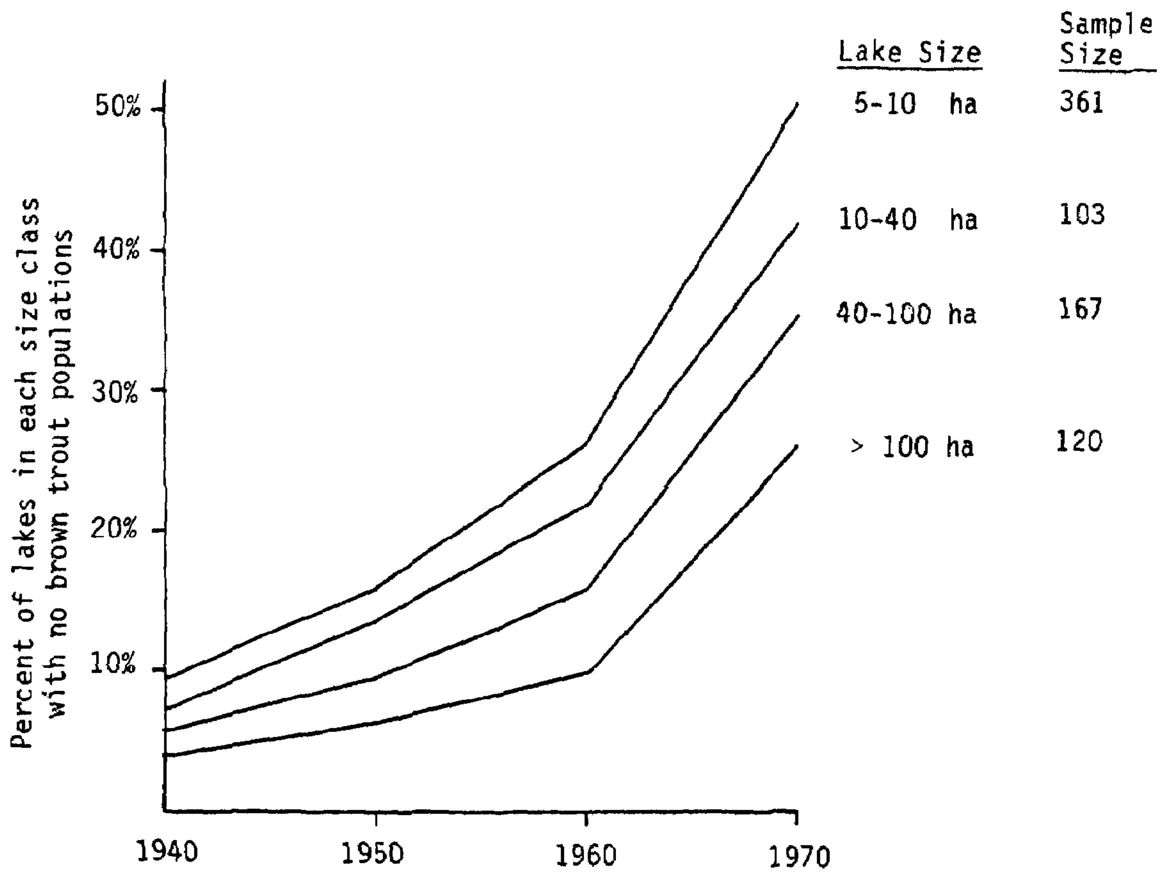


Figure 26. Extinction of brown trout populations in lakes of southern Norway during the period 1940-1970. Populations lost in each ten-year period are indicated for four different categories of lake size. (Adapted from Muniz and Leivestad 1980a)

status with pH, alkalinity, and aluminum concentrations in potentially sensitive regions; monitoring for fishkills, blood chemistry stress and changes in growth, age-class composition and reproductive success will permit the determination of trends in aquatic impact over time (Harvey 1980). Kelso et al. (1981) suggest that subtle qualities of surface water chemistry, biotic lifestyle and habitat modification must be considered in a complete assessment of biological effects.

5.3.6 Aquatic Ecosystem Response

The effects of acid deposition in aquatic ecosystems are numerous, complex, varied, and highly interrelated. In regions sensitive to acid inputs, virtually every aspect of ecosystem structure and function is subject to alteration. The nature of ecosystem damage in many ways reflects the variety and severity of responses observed in plant and animal populations. At the same time, the extent or rate of ecosystem degradation is intimately linked to the physico-chemical qualities characterizing lakes, rivers, and streams of different regions and continents. Effects may thus be anticipated to result from a combination of acid-induced changes in water chemistry and biotic composition. Much of the current state of knowledge has been gained through long-term interdisciplinary research initiated in Norway. Research currently underway in eastern North America has confirmed many findings of the Scandinavian works, but regional differences demand a great deal of care in applying the Norwegian experience to North America. Also, many complex issues remain to be resolved among the wide variety of scientific disciplines contributing to studies of aquatic ecosystem-level impacts.

Freshwater acidification has been demonstrated to alter nutrient cycling in streams and lakes by suppressing organic litter decomposition. As water pH levels decline, the activity of aerobic bacteria is diminished and functions of decomposition and mineralization are gradually taken over by less efficient fungi (Grahn et al. 1974; Hultberg and Grahn 1976; Likens 1976). Reductions in microbial biomass have been measured by declines in weight loss of leaves in litterbags placed in acid lakes (Hendrey et al. 1976); depressed community respiration has also been shown to occur in response to increased acidity (Traaen 1980). As a result of depressed microbial activity, organic litter accumulates at an accelerated rate and fewer nutrients are made available to primary producers (Friberg et al. 1980; Gahnstrom et al. 1980; Hall and Likens 1980; Hendrey and Vertucci 1980). These effects may be particularly pronounced in streams, where allochthonous material is the most important nutrient source (Traaen 1980).

Nutrients in the water column can be further reduced by interactions with metals in precipitation and surface run-off. Phosphorus and possibly silica are precipitated to the sediments by aluminum and iron ions present in waters of low pH (Dickson 1980). Organic carbon may be lost in a similar fashion (Muniz and Leivestad 1980a). Work in terrestrial ecosystem response also suggests that watershed acidification may result in reduced exports of phosphorus and other nutrients to aquatic systems (Hendrey et al. 1980a).

The effects of acidification on nutrient cycling are stimulated by the spread of periphyton and aquatic moss to deeper substrates. These plants form thick mats which effectively seal off the sediments from chemical exchange with the water column, inhibiting the release of sediment-bound nutrients and detritus (Grahn et al. 1974; Hendrey and Vertucci 1980). Anaerobic conditions created in upper sediment layers suppress bacterial and invertebrate populations alike while promoting the establishment of odor-producing, anaerobic bacteria (Hendrey 1979). Sediment aeration is further inhibited by reductions of rooted macrophyte communities (Leivestad et al. 1976).

The aquatic mosses, especially Sphagnum, may also contribute to freshwater acidity and oligotrophication (Grahn et al. 1974; Grahn 1977; Hendrey and Vertucci 1980). Once established, these acidophilic plants are believed to accelerate the acidification process by giving off hydrogen ions in exchange for available cations in the water column. This process is enhanced by the abundance of trace metal ions in acid lakes, however essential elements such as calcium and potassium are also readily sorbed by Sphagnum. The result is a combination of increased acidity, reduced buffering capacity, and diminished fertility which together contribute to biotic impact and accelerate the oligotrophication of acid lakes. A simplified flow diagram encompassing major portions of the overall process is presented in Figure 27. The Sphagnum invasion and its chain of effects are thought to be initiated by the acid pulse of spring snowmelt, when competing benthic flora and fauna are weakened or only partially established (Hultberg 1977).

Energy flows between trophic levels may be disrupted or reduced when primary productivity declines, or when variations occur in the relative productivity of competitive species. As discussed above, sensitive fauna may expend additional energy counteracting physiological stress, to the detriment of their growth and successful reproduction. Food chains are further altered and simplified through the elimination of some species and decreased numbers of others. Benthic detritivores, for example, are known to prefer to feed on detritus conditioned by bacteria rather than the relatively cruder by-products of fungal decomposition; many also derive a large portion of their energy from the bacteria contained in detritus (Hendrey et al. 1976). The reduction of bacterial populations accompanying oligotrophication would adversely affect these detritivores.

Populations of invertebrates are further reduced by the loss of habitat accompanying Sphagnum invasion (Grahn et al. 1974), potentially affecting the variety and amount of food available to fish, waterfowl and

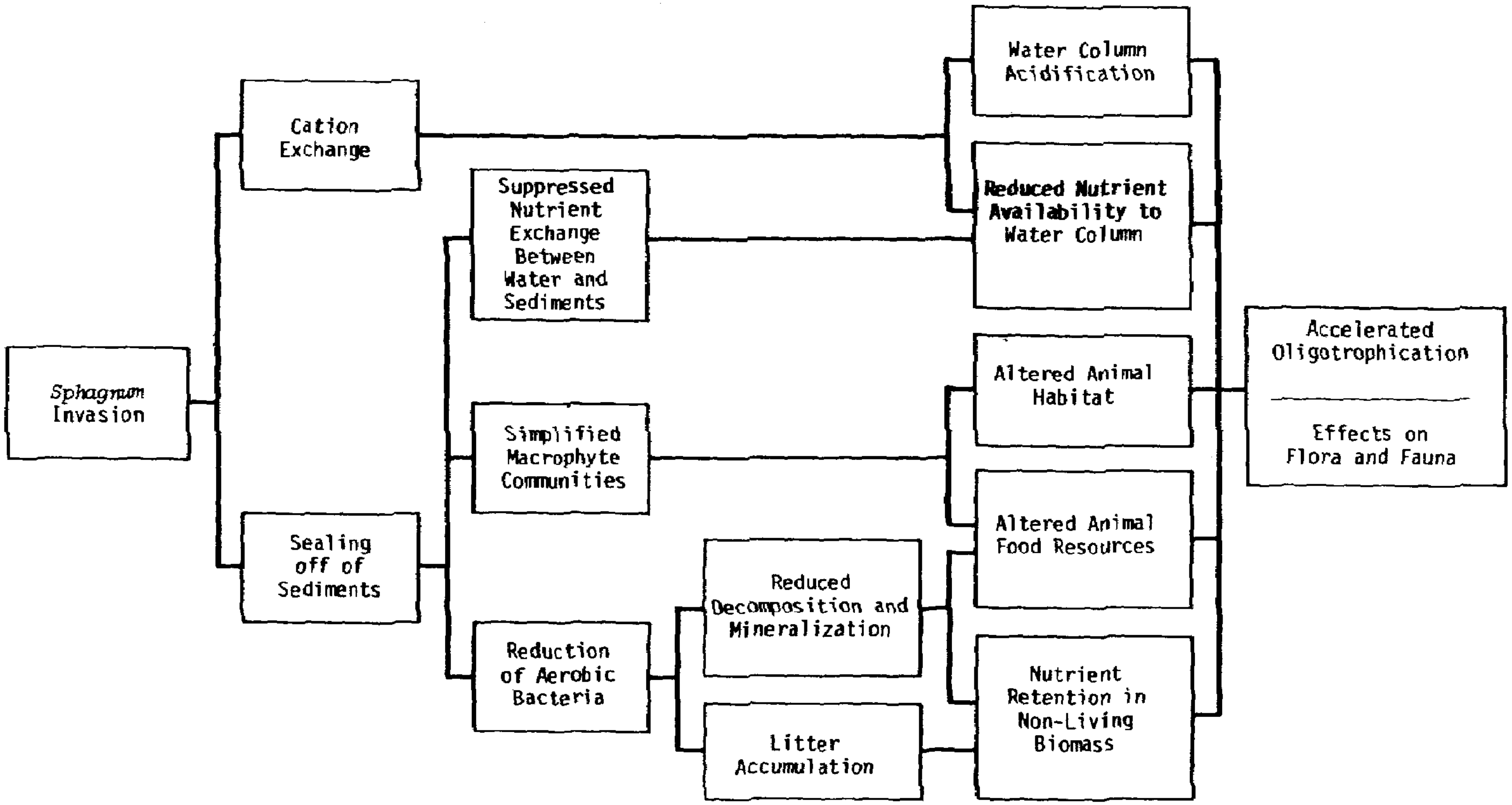


Figure 27. A schematic representation of the hypothesis of auto-oligotrophication in acid lakes.

other consumers at higher trophic levels (Hendrey 1978). The elimination of fish populations, largely through alterations of prey-predator relationships, can also induce radical changes in energy transfer along the food chain (Friberg et al. 1980; Henriksen et al. 1980; Muniz and Leivestad 1980a).

In sum, the effects of acid deposition on aquatic systems are cumulative and they interact in a number of ways to compromise ecosystem structure and function. Successional processes that promote the gradual eutrophication of surface waters may be interrupted, causing the systems to regress through less mature evolutionary stages to an increasingly oligotrophic state. The most pronounced effects will involve:

- reductions of nutrient cycling;
- disruptions of trophic relationships;
- alterations of the spatial and temporal distribution of species; and
- arrested ecosystem evolution.

The long-term consequences of large-scale alterations are difficult to predict with any certainty and the potential reversibility of these effects in aquatic ecosystems remains unknown.

6.0 AIR QUALITY LEGISLATION

The most important piece of Federal Legislation dealing with air pollution is the Clean Air Act, as amended in 1977 (42 USC 7401 et seq.). In this chapter, the main features of the law are described. It should be recognized that this discussion is by no means a complete presentation of all aspects of this very complicated law. For additional details the reader should refer to the summaries in Arbuckle et al. (1979), Avery and Schreiber (1979), and Quarles (1979). This chapter also describes the Acid Precipitation Act of 1980 as well as other activities undertaken to deal with the international problems associated with the long-range transport of air pollutants. In specific cases local environmental regulations, which are not discussed in this report, must also be reviewed, since they may be more stringent than Federal and State regulations.

6.1 THE CLEAN AIR ACT

The principal features of the Clean Air Act are the requirements for the Federal government to set standards for pollution levels and for the states to develop State Implementation Plans (SIPs) to meet the standards. Acronyms representing the major structural elements of this legislation are provided in Table 29. A graphic overview of the Clean Air Act is presented in Figure 28.

6.1.1 Federal Air Quality Standards

The Clean Air Act, as amended, embodies Federal efforts to establish maximum permissible concentrations of major air pollutants throughout the entire United States. National Ambient Air Quality Standards (NAAQS) are based on pollutant concentration per volume of air (ug/m^3 or mg/m^3) and have been set for six criteria pollutants: sulfur dioxide (SO_2), nitrogen dioxide (NO_2), carbon monoxide (CO), ozone (O_3), hydrocarbons (HC) and particulates. A comparable NAAQS has also been formulated for lead (Pb).

The primary ambient air quality standards specify atmospheric concentrations of the criteria pollutants which, if surpassed, could adversely affect human health. Secondary ambient air quality standards specify those ambient concentrations which, if exceeded, could have deleterious effects on public welfare. The concept of public welfare includes living resources such as fish and wildlife, and their habitats.

Both long-term and short-term NAAQS have been set. The former specify pollutant concentrations which cannot be surpassed on an annual average, and the latter dictate levels not to be exceeded over a period ranging from three to twenty-four hours. These standards are based on criteria documents which collect the scientific evidence regarding the effects of air pollution on health and welfare. Table 30 presents the primary and secondary NAAQS. While these standards are relevant to ground-level air pollution control, they do not directly address problems of atmospheric deposition in sensitive ecosystems.

Table 29. Acronyms for principal components of the Clean Air Act.

AQCR	Air Quality Control Regions
BACT	Best Available Control Technology
LAER	Lowest Achievable Emission Rate
NAAQS	National Ambient Air Quality Standards
NESHAPS	National Emission Standards for Hazardous Air Pollutants
NSPS	New Source Performance Standards
PSD	Prevention of Significant Deterioration
SIP	State Implementation Plan

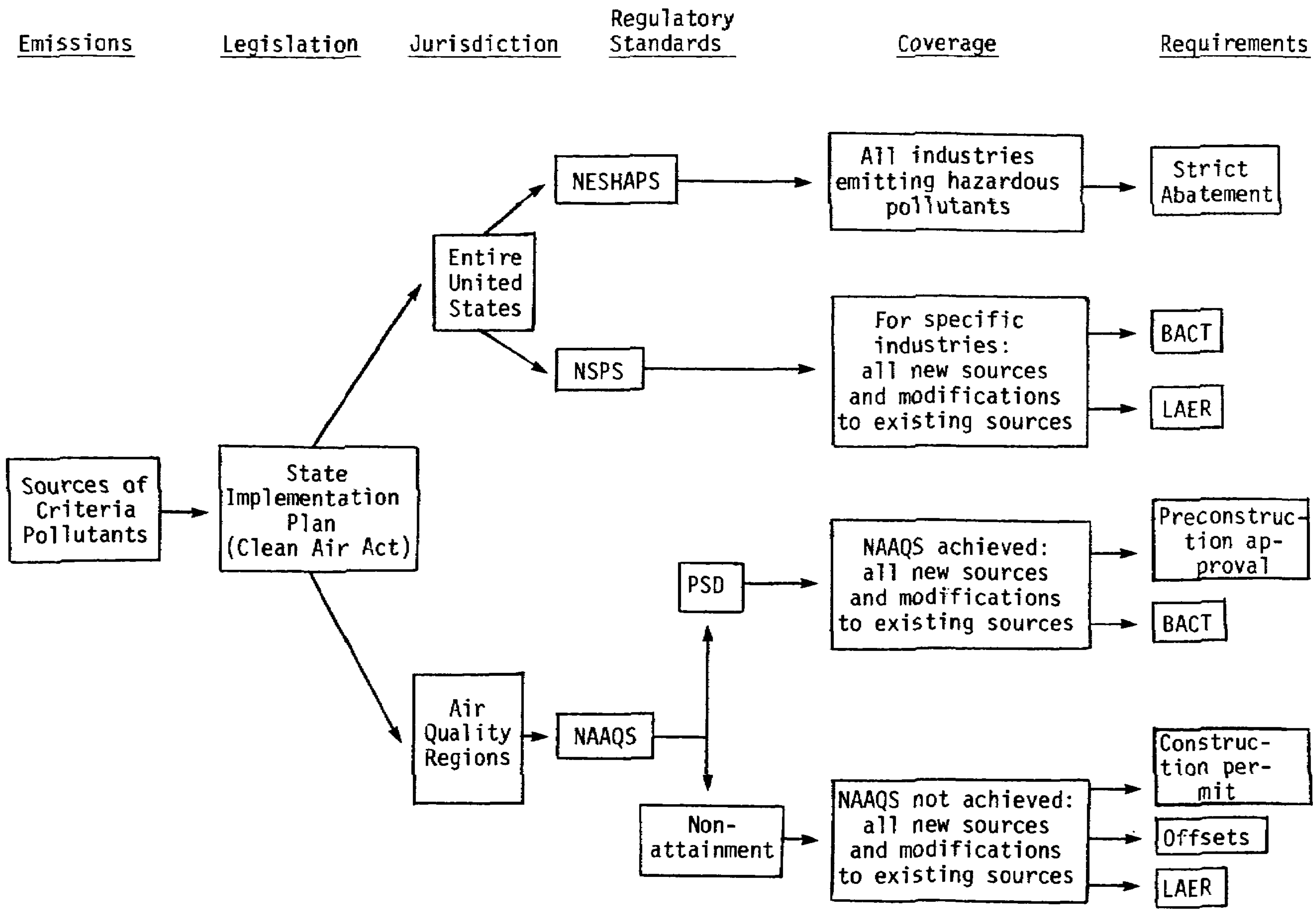


Figure 28. An overview of the Clean Air Act.

Table 30. National primary and secondary ambient air quality standards.

Pollutant	Air Quality Standards (micrograms/m^3)			
	Primary		Secondary	
	Annual Mean	Maximum Concentration (Allowed Once Yearly)	Annual Mean	Maximum Concentration (Allowed Once Yearly)
Sulfur Oxides (SO_X) (measured as SO_2)	80	365 (during 24 hours)	--	1300 (during 3 hours)
Particulates	75	260 (during 24 hours)	60	150 (during 24 hours)
Carbon Monoxide (CO)	--	10 milligrams/m^3 (during 8 hours)		
		40 milligrams/m^3 (during 1 hour)		Same as Primary Standard
Ozone (O_3)	--	240 (during 1 hour)		Same as Primary Standard
Hydrocarbons (HC)	--	160 (during 3 hours 6:00-9:00 a.m.)		Same as Primary Standard
Nitrogen Dioxide (NO_2)	100	---		Same as Primary Standard
Lead	--	1.5 (Averaged over calendar quarter)		Same as Primary Stantard

(From Council on Environmental Quality 1980)

6.1.2 Federal Emission Standards

The Clean Air Act directs EPA to establish New Source Performance Standards (NSPS) for emissions of criteria pollutants which apply to selected industries. The owner of a proposed major source must notify the state within which the source will be located before beginning construction or operation. The levels set by EPA are based on the best demonstrated technology for controlling emissions, taking into account economic feasibility.

No construction permit which the act requires can be issued for a source in any of the specified categories unless the emissions will meet the NSPS standards. However, the 1977 Amendments to the law contain additional regulations which must be incorporated into the SIPs to control emissions from new sources and modifications to existing sources. These approaches impose emissions limitations which are more stringent than the NSPS. The NSPS is therefore the upper limit for emissions from new sources in the specified industries.

Other emission standards under the law deal with hazardous air pollutants. The National Emission Standards for Hazardous Air Pollutants (NESHAPS) have been adopted to control asbestos, beryllium (Be), mercury (Hg), and vinyl chloride. These emission standards are based strictly on human health considerations. Unlike the NSPS, the NESHAPS apply to both new and existing sources of the hazardous pollutants.

Title II of the Clean Air Act, also known as the National Emissions Standards Act, placed emission limitations on mobile sources of ambient air pollution. Mobile emissions are measured in grams of pollutant per vehicle mile over the lifetime of the source. For legal purposes, emission limits are expressed by emission factors, also measured in grams per mile, as shown in Table 31. These factors are often lower than actual emission limits due to potential emission increases associated with the deterioration of the vehicle.

6.1.3 State Responsibilities Under the Clean Air Act

The goal of Federal air pollution control legislation is to make enforcement activities the responsibility of the States. Under the Clean Air Act, the States are directed to establish State Implementation Plans (SIPs) which embody the necessary abatement procedures required to attain the NAAQS. However, if the State and local governments are unable to develop an acceptable SIP, the EPA is directed to impose the necessary regulations.

The 1970 legislation required that the country be divided into Air Quality Control Regions (AQCRs). These 247 regions are highly variable in size and independent of jurisdictional boundaries. Some are contained within state boundaries while others include portions of several states. The AQCRs were selected on the basis of regional homogenity in existing

Table 31. Mobile source exhaust emission factors for 1979, 1980 and 1985-1990.

	Emission factors (grams/mile)								
	Nitrogen Oxides			Hydrocarbons			Carbon Monoxide		
	1979	1980	1985-1990	1979	1980	1985-1990	1979	1980	1985-1990
Automobiles	1.5	1.5	0.29	1.13	0.13	0.13	18.6	3.0	1.4
Light duty gasoline trucks	1.73	1.73	0.41	0.94	0.94	0.31	14.5	14.5	3.87
Heavy duty gasoline trucks	9.1	9.1	4.0	5.22	5.22	1.46	191.9	191.9	15.4
Heavy duty diesel trucks	19.9	19.9	5.35	4.5	4.5	2.85	27.0	27.0	27.0

(Adapted from MITRE 1978)

air quality data, urban concentrations, types of industry and nature of emissions, terrain, and meteorological conditions (Arbuckle et al. 1979; Quarles 1979). When an AQCR is found to be in violation of the NAAQS, the SIP must delineate a combination of control strategies intended to bring the AQCR into compliance.

Primary standards are to be attained "as expeditiously as practicable" and secondary standards within a "reasonable time". Each SIP contains:

- a description of the air quality in each AQCR;
- an inventory of emission sources;

- emissions limitations and compliance schedules for each source;
- a permit program for new source construction; and
- procedures for monitoring, reporting, and enforcement.

The 1977 Amendments required the states to designate areas which meet the NAAQS (attainment areas) and those which do not (nonattainment areas). Because the AQCRs are defined without regard to political boundaries and cross state lines, the States have not used AQCRs in defining attainment and nonattainment areas. As a result, the focus of activity under the Clean Air Act has shifted to the regulation of sources in attainment and nonattainment areas, reducing the significance of the AQCRs (Arbuckle et al. 1979).

a. Nonattainment areas. The SIPs must incorporate the NSPS discussed above, ensuring that all new sources meet these standards. In addition the SIPS must describe the mechanism for review of new sources in areas not meeting the NAAQS (nonattainment areas). Any new source or modification of an existing source with potential emissions of 100 tons per year or more of particulates, SO_2, NO_x, volatile organic compounds, or carbon monoxide is subject to a construction permit requirement. In nonattainment areas, building and operating a major source requires the control of pollutants to the Lowest Achievable Emission Rate (LAER) for a given industrial category. The LAER will always be at least as stringent as the NSPS. For construction and operation permits to be issued the following conditions must be met:

- all other installations in the state of the same ownership must be in compliance, or on an approved schedule to achieve compliance, with the abatement portions of the SIP;
- any projected new emissions must be more than equally offset by reductions from existing sources of air pollution in such a way as to produce "a positive net air quality benefit" in the area; and
- new sources in nonattainment areas must meet the LAER defined for each case.

b. Attainment areas. The Clean Air Act Amendments of 1977 provide the legislative basis for actions to maintain air quality in areas where ambient standards are already being met (attainment areas). The amendments for the most part adopted regulations which had been promulgated earlier by EPA, incorporating them into Part C (Sections 160-169) of the Act. These Sections establish the program to Prevent Significant Deterioration of Air Quality (PSD). Under Section 161 the SIPs must be revised to include any measures needed to prevent the degradation of air quality in attainment areas, and Section 168 specifies that until the SIPs are modified to meet this requirement, the regulations issued earlier by EPA will remain in force.

As was the case for nonattainment areas, a preconstruction review of major sources and modifications to existing sources in attainment areas must also be made under the PSD program (Quarles 1979). The PSD program applies to stationary sources in a specified set of industrial categories that will emit at least 100 tons per year of any regulated air pollutant. Sources not included in the specified industrial categories are also covered if their emissions will exceed 250 tons per year. Sources located in nonattainment areas also fall within the scope of the PSD program if their emissions will effect attainment areas.

Within the PSD program, areas of the country which meet the NAAQS are divided into three classes: Class I for areas of restricted growth, Class II for areas of moderate growth, and Class III for industrialized areas. Permissible increases in ambient pollution levels depend on the designated area classification, but in all cases those levels must remain below the NAAQS. New plant construction approvals depend on the classification of the affected area, the resulting increase in air pollutant concentration, and a general requirement to employ the Best Available Control Technology (BACT), as determined by the permitting authority for each case (Quarles 1979). BACT may not be less stringent than NSPS. Maximum increases in concentrations of SO_2 and particulates have been promulgated for each class of area and are shown in Table 32.

In addition to the requirements for BACT and the specification of maximum allowable increments, the applicant for a preconstruction permit must provide the results of an analysis of the impact on soils, vegetation, and visibility of the proposed source and any associated development. There is also to be a public hearing. The applicant must provide air quality monitoring data for a period of one year prior to construction, and agree to monitor air quality after operation begins.

c. Review by federal officials. Section 165 of the Act provides that, in Class I areas, Federal Land Managers (FLM) are responsible for assuring that potential emissions in and around areas under their jurisdiction will not contribute to the deterioration of "air quality related values (including visibility)" of these lands, where Federal Land Manager means the Secretary of the department with authority over the land. If the Federal official charged with direct responsibility for management of any lands within a Class I area or the Federal Land Manager of such lands finds that the new facility will have an adverse effect on air quality related values, then the permit may be denied, even if the increase in pollutant concentrations is less than the maximum allowable increase for a Class I area. The owner of the proposed facility can appeal to the Governor who may recommend a variance, and the permit may be issued if the FLM agrees. However, in cases where the Governor recommends a variance in which the FLM does not concur, their recommendations go to the President for a final decision. If the increase in pollutant concentration exceeds the maximum allowable for Class I areas, a permit may still be granted if the FLM certifies that no adverse impact on air quality related values will occur.

Table 32. Prevention of significant deterioration regulations. Maximum allowable increase over baseline concentrations.

Pollutant	Land Classification		
	Class I	Class II	Class III
Sulfur dioxide (micrograms/m^3)			
Annual geometric mean	2	20	40
24 hour maximum	5	91	182
3 hour maximum	25	512	700
Particulates (micrograms/m^3)			
Annual geometric mean	5	19	37
24 hour maximum	10	37	75

(From Avery and Schreiber 1979)

It is possible for an area to be classified attainment for certain pollutants and nonattainment for others. In addition, pollutants from a proposed plant in an attainment area may cross boundaries into a nonattainment area. Thus the PSD and nonattainment regulations may overlap (Quarles 1979). The provisions of a SIP constitute a highly complex regulatory process which varies from state to state. Moreover, area designations are subject to periodic revision and careful attention is therefore required to assure that the most up-to-date versions are employed in the regulatory activities of Federal agencies.

6.1.4 Discussion

The rationale for Federal control of air pollutant emissions is based, at least partially, on the transboundary nature of the long-range transport of air pollution and on the inability of any state to control pollution which is generated outside its borders. Pollution from midwestern states, for example, is transported to states in the northeast, possibly using up their PSD increments or rendering areas nonattainment. This in effect would restrict the potential for economic development in the receptor states. The impacts would also reduce the economic value of wildlife resources used for recreational purposes.

Activities under a SIP are designed to achieve and maintain satisfactory air quality within that state. The transport of air pollution across state borders will contribute to the deterioration of air quality in receptor states, and stricter emission controls will then be required on sources in those states. The rigid pollution controls required for northeastern development have prompted lawsuits by some of these states to enforce similarly stringent controls of emissions in source states. The strengthening of midwestern SIPs would, in turn, impact the economies of those states by increasing pollution control costs.

Effective regulatory control of acid precipitation in the future may directly involve both the NAAQS and performance standards. Emissions standards for industries emitting acid precursors should result in a reduction of both total emissions and atmospheric loadings of sulfur and nitrogen compounds through the use of control devices or fuels which have been cleansed of their sulfur. The recent NSPS for coal-fired power plants are a positive step towards decreased total SO_2 emissions in the eastern United States (Wetstone 1980). However, a revised set of primary NAAQS based on more than strictly health-related considerations would require additional legislation. Such legislation would require interdisciplinary research, quantified scientific evidence, and consistent public testimony relative to the subtle ecological effects of atmospheric deposition (Berry and Bachmann 1977).

6.2 THE ACID PRECIPITATION ACT

A recent piece of Federal legislation directed at limiting air pollution effects on wildlife and habitat resources is the Acid Precipitation Act of 1980 (42 USC 8901 et seq.). This act mandates the formation of

Table 33. Federal departments and agencies participating in the Interagency Task Force on Acid Precipitation.

DOA	Department of Agriculture co-chair
EPA	Environmental Protection Agency co-chair
NOAA	National Oceanic & Atmospheric Administration co-chair
CEQ	Council on Environmental Quality
DOC	Department of Commerce
DOE	Department of Energy
DOI	Department of Interior
DOS	Department of State
DHHS	Department of Health & Human Services
NASA	National Aeronautics and Space Administration
NSF	National Science Foundation
TVA	Tennessee Valley Authority

(From Interagency Task Force on Acid Precipitation 1981) the Interagency Task Force on Acid Precipitation. Composed of DOI, EPA, and ten other Federal departments and agencies (Table 33), this group is charged with the development of a ten-year research program, the National Acid Precipitation Assessment Plan (Interagency Task Force on Acid Precipitation 1981).

6.3 INTERNATIONAL COOPERATION

Since the 1972 United Nations Conference on the Human Environment in Stockholm, the European Economic Community (EEC), the United Nations (UN), and the Organization for Economic Cooperation and Development (OECD) have all undertaken efforts to create guideines for international programs for environmental protection. The long-range transport of air pollution has been the subject of numerous ethical statements from international organizations as well as proposed treaties in northern Europe. The first international agreement on the long-range transport of air pollutants entered into by the United States was the "Convention on Transboundary Air Pollution" of the Economic Commission for Europe, a UN organization. While it provides no abatement schedule or other enforcement provisions, it develops routes of international cooperation in research relevant to transboundary air pollution and its socioeconomic and ecological repercussions. The United States and Canada have signed a "Memorandum of Intent on Transboundary Air Pollution" for an intensive research program leading to the preparation of bilateral agreement (External Affairs Canada 1980). The U.S.-Canada border crosses large, isolated areas which are well suited for the observation of transboundary pollution and its effects. This agreement therefore could serve as a model for future international agreements (Wetstone 1980).

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GLOSSARY

acid - A substance which can donate hydrogen ions.

acid rain - A popular term for acid precipitation, or rain principally containing the hydrolyzed end-products of oxidized sulfur and nitrogen substances (dilute strong acids) and of pH less than 5.6, the minimum pH expected from equilibrium with atmospheric carbon dioxide.

advection - The horizontal movement of an air mass due to atmospheric pressure gradients.

adsorption - Adhesion of a thin layer of molecules to a liquid or solid surface.

aerodynamic diameter - An expression of the aerodynamic behavior of an irregularly shaped particle in terms of the diameter of a sphere of unit density having identical aerodynamic behavior to the particle in question.

aerosol - Solid particles or liquid droplets which are dispersed or suspended in air.

air quality standards - Concentrations of air pollutants which cannot legally be exceeded during fixed time intervals within specified geographic areas.

alkalinity (total) - A measure of the concentration of all acid-neutralizing substances in solution.

ambient air - Air surrounding a given point.

anion - A negatively charged atom or molecule.

antagonism - A relationship in which the combined action or effect of two or more pollutants is less than the sum of their individual effects.

area source - A geographic location from which pollutants are emitted and transported by advection.

bioindicator - A plant or animal species sufficiently sensitive to a given pollutant to be useful as an indicator of the presence of the same pollutant.

buffer - A substance in solution capable of neutralizing both acids and bases, thereby maintaining the original pH of the solution.

buffering capacity - The ability of an entity (e.g., body of water or its watershed) to neutralize introduced acids.

catalyst - A substance capable of increasing the speed of chemical reactions without itself undergoing physical or chemical change.

cation - A positively charged atom or molecule.

cellular permeability - The ability of cells to absorb or evacuate substances across cell membranes; a sensitive indicator of injury to deep-lung cells.

dose - A measured concentration of a toxicant for a known time period during which a subject is exposed.

dry deposition - Matter transferred from the atmosphere to ground in the absence of precipitation; also the process of such transfer, including surface adsorption of gases, sedimentation, Brownian diffusion, and particle impaction.

dust - Solid particles generated by physical alterations of organic or inorganic substances.

ecosystems - The interacting system of a biological community and its environment.

effluent - Any solid, liquid, or gaseous waste emitted by a process.

emission standards - Standards based on the concentration of pollutants from stacks that cannot legally be exceeded during fixed time intervals within specified geographic areas.

fluorides - gases or particles containing fluorine compounds.

fly ash - Suspended incombustible or partially incinerated matter carried in the gaseous products of combustion.

fossil fuel - Fuel derived from decayed organic matter from past geologic ages.

fumigation - The natural or controlled exposure of biota to toxic gases or volatile substances.

haze - Fine dust, smoke or vapor which reduces the transparency of air.

heterogeneous process - A chemical reaction involving reactants of more than one phase or state, such as one in which gases are adsorbed to aerosol droplets where the reaction takes place.

hydrocarbons (HC) - Organic compounds of carbon and hydrogen derived mainly from fossil deposits and vegetative sources; along with NO_x, they are implicated in the formation of ozone and other photochemical oxi- dants in smog.

hydrometeor - A product of the condensation of atmospheric water vapor.

impaction - An impinging or striking of one object against another; also, the force transmitted by this act.

inversion - A thermally stable atmospheric condition wherein cooler air is prevented from rising by a layer of warmer air above.

isopleth - A line on a map connecting points at which a given variable has a specified constant value.

leach - To dissolve out through the action of a percolating liquid.

LRTAP - The long-range transport of air pollution, often analagous to transboundary air pollution.

mass median diameter (MMD) - The geometric median size of a distribution of particles based on weight.

mobile source - A moving source of air emissions.

monitoring - The use of gas sensing instruments or other devices to measure the concentrations of pollutants.

mutagen - Any agent that induces inheritable genetic change in living organisms.

nitrogen oxides (NO_x) - A class of nitrogen and oxygen compounds which includes nitric oxide (NO), nitrogen dioxide (NO_2), and nitrogen trioxide (NO_3); derived from many natural sources as well as fossil fuel combustion, they participate with hydrocarbons in the formation of photochemical smog, and with sulfur oxides in causing acid precipitation.

oxidant - A chemical compound which has the ability to remove electrons from another chemical species, thereby oxidizing it; also, a substance containing oxygen which reacts in air to produce a new substance, or one formed by the action of sunlight on oxides of nitrogen and hydrocarbons.

ozone (O_3) - A colorless to faintly bluish, unstable, pungent gas produced by electrical discharge in air, by solar ultra-violet radiation, or by other photochemical reactions of mixtures of certain hydrocarbons and nitrogen oxides; a strong oxidizing agent that is phytotoxic at low concentrations.

PAN - The acronym for peroxyacetyl nitrate ($CH_3C\text{-}O\text{-}NO_2$); the principal constituent in a homologous series of compounds, referred to as peroxyacyl nitrates or PANs, and formed as a product of photochemical reactions involving nitrogen dioxide and hydrocarbons.

particulates - Fine liquid or solid particles, such as dust, smoke, mist fumes or smog, found in the air or in atmospheric emissions.

pathogen - Any biotic or abiotic agent capable of causing disease.

photochemical oxidants - Primarily ozone, nitrogen dioxide and PAN, along with lesser amounts of other compounds, formed as products of atmospheric reactions involving organic pollutants, nitrogen oxides, oxygen and sunlight.

plume - The path taken by pollutants emitted continuously from a point or area source.

podzol - Any of a group of zonal soils that develop in a moist climate, especially under coniferous or mixed forest; they are characteristically acidic and low in essential plant nutrients.

point source - A stationary emitting point of air pollution.

pollutant (air) - Any gas, liquid, or solid contaminant present in the atmosphere in such quantity as to cause undesirable effects on living organisms or materials.

precipitation scavenging - The capture of air pollutants within (rainout) and beneath (washout) clouds by a hydrometeor.

primary pollutants - Pollutants which are emitted directly from an identifiable source.

rainout - Removal of particles and/or gases from the atmosphere by their involvement in cloud formation (particles act as condensation nuclei, gases are adsorbed by cloud droplets), with subsequent precipitation.

secondary pollutants - Pollutants produced in the air by reactions involving primary pollutants and/or other atmospheric constituents.

sink - A reactant with or absorber of another substance.

smog (general) - a mixture of smoke and fog.

(London type) - A mixture of coal smoke and fog, with sufficient sulfur dioxide to induce chemical reducing properties.

(Los Angeles type) - A mixture of photochemical oxidants, with sufficient ozone to induce chemical oxidizing properties.

sulfur oxides (SO_x) - A class of sulfur and oxygen compounds composed primarily of sulfur dioxide (SO_2) and sulfur trioxide (SO_3); they are derived mainly from anthropogenic sources and participate with nitrogen oxides in the formation of acid precipitation.

synergism - A relationship in which the combined action or effect of two or more pollutants is greater than the sum of the effects of the individual pollutants.

total suspended particulates (TSP) - An aggregate measure of solid and liquid particles present in the atmosphere; it may contain toxic trace substances and interact synergistically with sulfur oxides.

toxicant - A substance that kills or injures living organisms by its chemical or physical action, or by altering the environment of the organism.

trace elements - Atomic species suspended in air at natural concentrations of less than 1.0 ppm; toxic and possibly carcinogenic elements at low concentrations include beryllium, cadmium, fluorine, lead, manganese, mercury and selenium.

washout - The capture of gases and particles by hydrometers falling beneath clouds.

wet deposition - Matter transferred from the atmosphere to ground in precipitation; also the process of such transfer.

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This introductory volume synthesizes results of scientific research related to air pollution effects on fish and wildlife resources. It is intended for use as a general reference to provide background information for the 8 ecosystem specific reports in this series: Deserts and Steppes, Forests, Grasslands, Lakes, Rivers and Streams, Tundra and Alpine Meadows, Urban Ecosystems, and Critical Habitats of Threatened and Endangered Species.

Related air pollutants are classified in three categories. A general summary of pollutant origins, atmospheric transport, transformation and deposition is presented. The report describes plant, animal and ecosystem responses to air pollution as well as factors affecting the sensitivity of receptive ecosystems. This volume also briefly describes relevant features of air quality legislation.

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